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MODERN METHODS FOR ESTIMATION OF PARTICLE DENSITY

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ABSTRACT

Particle density, N_V, or more generally the density of 3-dimensional objects such as grains in polycrystals and pores in sintered materials, is one of the basic structural parameters used in materials science. Methods for estimation of particle density are perceived by a large fraction of the materials science community as complicated and time consuming. The aim of the present paper is to show that the negative perception of procedures for N_V estimation slowly looses its basis due to the progress made in specimen preparation, imaging techniques and dissemination of high power microcomputers.

Key words: disector, particle density.

INTRODUCTION

Particle density, N_V, or more generally the density of 3-dimensional objects such as grains in polycrystals and pores in sintered materials, is one of the basic structural parameters used in materials science. Its importance is illustrated in particular by current models of particle and grain size strengthening: e.g. (Armstrong et al., 1962; Ralph and Hansen, 1982; Rhines, 1986; Kurzydłowski and Bucki, 1993). Despite that, estimates of N_V are rarely found in the literature. This only to some extent can be attributed to insufficient knowledge of stereological methods among materials science specialists. The major obstacle for N_V becoming a standard characteristic of materials has been in the past the complexity of its measurement.

Methods for estimation of particle density are perceived by a large fraction of materials science community as complicated and time consuming. Such a perception is supported by the fact that unless the specimens are made extremely thin then most materials are opaque to illumination by electrons, light and other electromagnetic waves which are widely utilized in microscopic methods. The other factor is disappointment related to the fact that an intuition driven counting of particle sections revealed on cross-sections of materials in most cases does not yield a correct estimation of N_V.

The aim of the present paper is to show that the negative perception of procedures for N_V estimation slowly looses its basis due to the progress made in specimen preparation, imaging techniques and dissemination of high power microcomputers. At the same time the stereological complexity of the measurements remains intact: estimation of N_V requires „3-dimensional information" which can be obtained only from „in-depth" (or in other words „volumetric") observations of the materials microstructures.

The following text provides a short theoretical analysis of the problem. However, it concentrates attention on the practical aspects of measurements. The paper gives also examples of estimation of particle density which include the disector method and serial sectioning. The last part addresses the problem of density of grains in polycrystalline materials.

ANALYSIS

Generally, measurements of the density of particles (and other 3-dimensional objects) can be carried out either in a direct or indirect way. This present paper, which is part of a series of articles on stereological methods, discusses the direct methods which provide estimation of N_V directly from counting and/or geometrical measurements on images of microstructure.

(The indirect methods for estimation of N_V are based on the following relationship between the density of particles and their mean volume, E(V):

$$N_V = V_V / E(V), \tag{1}$$

where V_V is the particle volume fraction. This relationship makes it possible to estimate N_V from a whole range of methods which provide estimation of the mean volume, E(V), recently reviewed for example in Gundersen (1986), Kurzydłowski and Ralph (1995)).

DIRECT MEASUREMENTS OF PARTICLE DENSITY

Direct measurements of particle density are based on an extremely simple concept which can be summarized as follows: take a known volume of the material studied and count the number of particles which it contains. However, a proper implementation of this simple concept is not trivial and deserves a comment.

As mentioned earlier the direct procedures for estimation of particle density inherently involve 3-dimensional, or in other words „volumetric" analysis. As a result the recent progress in stereology, which brought about new methods for estimation of particle volume, has made less impact on the techniques for estimation of N_V. In the case of this parameter the improvement is mainly related to refining and formalizing the already known and quite explicit procedures. Accordingly, the present text rather addresses implementation details than the philosophy of the estimation routines. In particular the attention in the present papers will focus on the following aspects of estimation procedures:

- randomization of observation fields
- imaging of particles of diverse size
- ambiguity of assigning particles to the volume studied.

RANDOM SELECTION OF OBSERVATION FIELDS

The very first problem which needs to be solved in the procedures for estimation of the particle density is the question of random selection of samples, fields of observations and eventually images. Dimensions of particles in modern materials range from 10 nm to 100 μm. This implies the need for high magnifications in the observations of the microstructures and in turn relatively small sizes of samples and small areas covered by an individual observation field. In such small specimens and observation fields particles may appear as distributed in a non-homogeneous way and the number of particles counted in a given field/image may show a significant scatter. Consequently, N_V requires a large number of observation fields which, for un-biased estimation should be selected in a random way. A simple method for randomization of observation fields selection is based on the concept of studying a fraction, say 30%, of samples prepared or images taken. Another solution is random positioning of microscope stage.

IMAGING OF PARTICLES OF DIVERSE SIZE

One of the major problems related to estimation of particle density is the need for imaging objects of sizes which vary from the nano- to micro-scale. In most cases of practical importance the large particles appear in low numbers and the value of N_V is strongly influenced by the proper estimation of a usually large number of small particles which can only be explored under a high magnification. On the other hand each method of microstructural imaging is characterized by a specific resolution limit and the smallest particle may not be revealed in particular observation conditions. The consequences of such a situation should be carefully analyzed and accounted for in the estimation procedures.
Observations of particles in a materials science context are carried out either on images of cross-sections or projections of particle embedded in a thin slice of the studied material. In the first case (images of sections) the available information is basically of a 2-dimensional character. As a result no assumption - free procedure allows for a direct estimation of N_V from a system of separate sections of the material. On the other hand, as is discussed further, a system of parallel sections, two at least, allows for an unbiased estimation of particle density.
Projected images of particles, in principle, are sufficient for the estimation of particle density providing that the thickness of the slice penetrated by the illumination is known. However, in this instance a problem arises of the particles emerging on the slice surfaces.

ONE PARTICLE - ONE OBSERVATION FIELD

Estimation of particle density via direct counting of number of the objects of interest, N, in a studied volume of the material in question, V, requires that each particle can be assigned to one and only one test volume disregarding the complexity of particle size/shape, complications of specimens preparation and imaging techniques.
In practice, the decision whether, or not a given particle, v_i, can be counted as belonging to the studied sample of the material, V, (and not to any other sample prepared in the same experimental conditions), usually is carried out through the following steps:

1. assignment of a representative point, x_i, to the particle in question,
2. rejection of the particles which do not meet the condition $x_i \in V$.

The most natural realization of step 1 is to use a point of the lowest or highest distance from the surface of the specimen. Accordingly, particles counted as belonging to a studied volume are those which are placed <u>below</u> the surface of observation (for observations on sections) or <u>below</u> the surface of the examined thin slice. Since counting must be carried out for a finite volume of material, one has also to define the surface <u>above</u> which particles are accepted. In the case of projected images the obvious choice is the second surface of the thin slice. On the other hand observations on sections require a second section placed at a known, and small distance from the first one. This leads to the method described in the literature as „disector": e.g. (Sterio, 1984; Liu, 1993).
The concept of two sections, S_1 and S_2, is the essence of direct measurement of N_V. The density is estimated from the following simple formula:

$$N_V = N / (A\ t) \tag{2}$$

where:
N is the number of particles which were placed below S_1 and above S_2, A is the observation field area, t is the distance between S_1 and S_2.

The following text gives examples of materials science application of this simple formula in the case of light and electron (scanning and transmission) microscopy. The examples cover both systems of isolated particles and closely packed polycrystalline aggregates.

DISECTOR METHOD IN THE CASE OF SCANNING ELECTRON MICROSCOPY (SEM)

Figure 1 shows SEM images (back scattered electrons) of the microstructure of a ceramic composite. This composite contains particles of tungsten carbide (white phase) in the matrix of alumina. Images in Fig.1.a-d were obtained on 4 consecutive sections, P_1, P_2, P_3, P_4 schematically shown in Fig.2. The sections were prepared by polishing in a water suspension of Al_2O_3. The distances between the sections, t_i, were estimated from measurements of changes in the size of indentations made by a diamond nano-indenter as explained in Fig.3. Images revealed on sections were organized into 3 pairs: (P_i, P_{i+1}) with i=1,2,3. Images of each pair were compared and particles were identified which are sectioned by the P_{i+1} and are not cut by P_i. These particles, circled in Fig.1, were counted as assigned to the volume V_i defined by t_i and the size of observation field. For each pair an estimate of N_V was obtained from the formula:

$$(N_V)_i = n_i / V_i \ (i=1,2,3) \tag{3}$$

where n_i is the number of particles counted in volume V_i. The results obtained for the sections P_1-P_4 are given in Table 1. In the whole experiment 10 randomly selected stacks of 4 sections were studied. They yielded $N_V=0.063\mu m^{-3}$

Table 1. Results of particle/particle section counting for the system of particles shown on Fig.1a-d

<table>
<tr><th>Section</th><th>Number of particle sections</th><th colspan="3">Number of particles in the disector</th></tr>
<tr><td>P₁</td><td>79</td><td rowspan="2">5</td><td></td><td></td></tr>
<tr><td>P₂</td><td>82</td><td rowspan="2">5</td><td></td></tr>
<tr><td>P₃</td><td>80</td><td></td><td rowspan="2">8</td></tr>
<tr><td>P₄</td><td>81</td><td></td><td></td></tr>
</table>

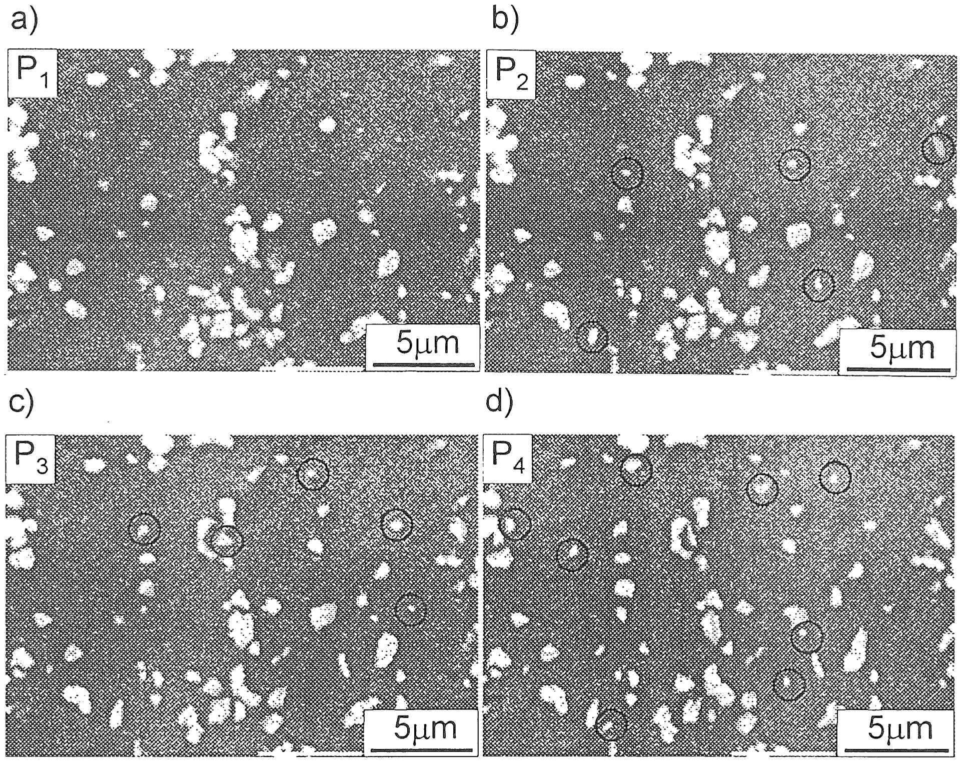

Fig.1. Set of parallel sections of particles of tungsten carbide.

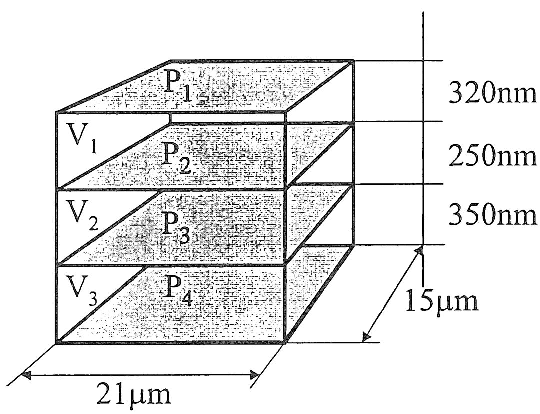

Fig.2. A schematic representation of a system of sections used for estimation of N_V.

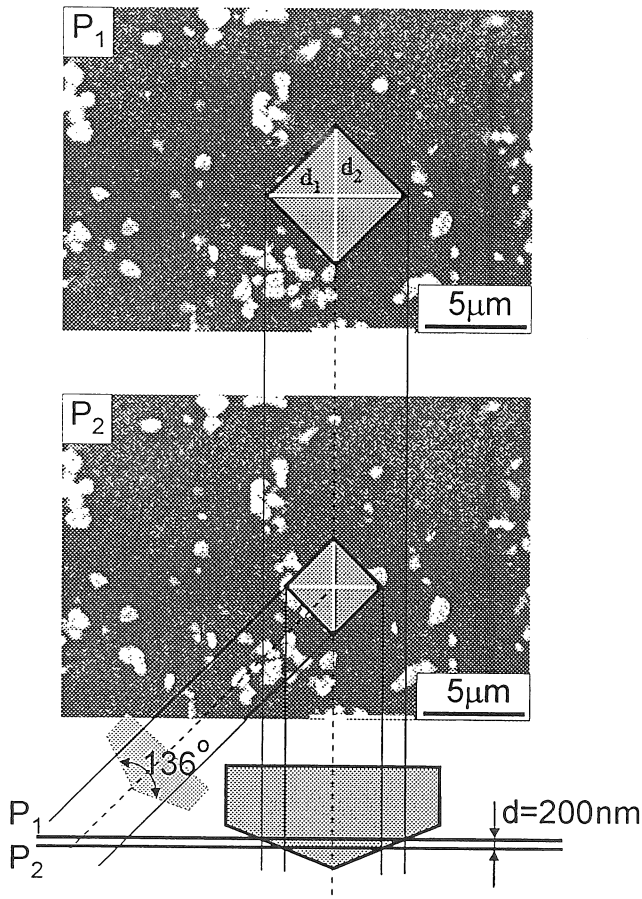

Fig.3. Illustration of the method used to estimate thickness of the layer of the material removed during polishing of the sample surface.

SERIAL SECTIONING

In the case of the disector method images obtained on a series of sections are organized in pairs which are analyzed in terms of the particles cut by only one of the section planes. With the recent progress in computer aided image analysis the examination can be extended and can take into account the position of each section of the particles. The images of these sections can be used subsequently for the reconstruction of the 3-D geometry of each particle. The procedure based on the analysis of series of sections is known in the literature as serial sectioning. An example of application of this method is shown in Fig.4 which presents the 3-D geometry of twin grains in a FCC metal (more details can be found in Bystrzycki et al., 1993).

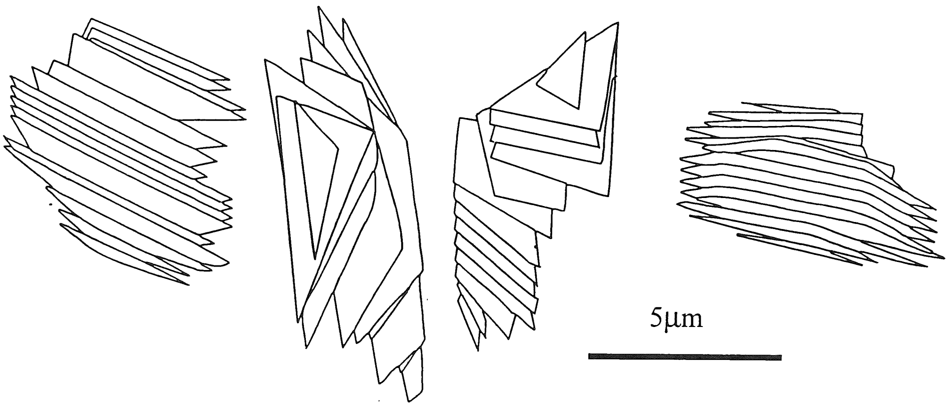

Fig.4. Examples of the geometry of twin grains in a FCC metal.

DISECTOR METHOD IN THE CASE OF TRANSMISSION ELECTRON MICROSCOPY (TEM)

Figure 5 shows a TEM image of carbides in an Fe-Cr alloy. This image is one of a series obtained on thin foils produced by ion milling. The thickness of the observation field (120 nm for Fig.5) was estimated for each observation field from the variation in the length of dislocation segments as a function of the angle between the electron beam and the normal to the thin foil.

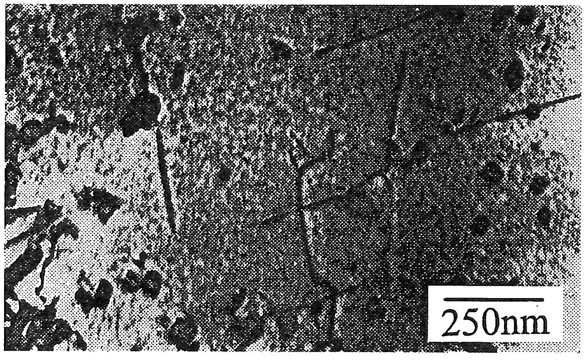

Fig.5. TEM image of carbides in FeCr alloy.

For each observation field the number of particle projections, n_i, was determined. However, as projected images do not easily differentiate images of particles emerging on the upper and lower foil surface, n_i cannot be directly used for estimation of N_V. In the present case a SEM mode was used to identify particles cut by upper surface of the thin foil. The number of such particles was subtracted from n_i and N_V was estimated using the same equation as in the previous example.

APPLICATION TO POLYCRYSTALLINE AGGREGATES

The tests were carried out on a pure, fully annealed iron which exemplifies a single phase polycrystalline material. The microstructures of cylindrical specimens were revealed on cross-sections parallel to their axis using standard metallographic procedures. The images of grain boundaries were digitized and transformed into binary ones (see Fig.6). These images were analyzed using an image analyzer. Grain area, A_i , and equivalent circle diameter, d_2, were measured for individual grain sections. The data in the form of the experimental distribution functions were characterized by their mean values E(x), standard deviations SD(x) and coefficient of variations CV(x).

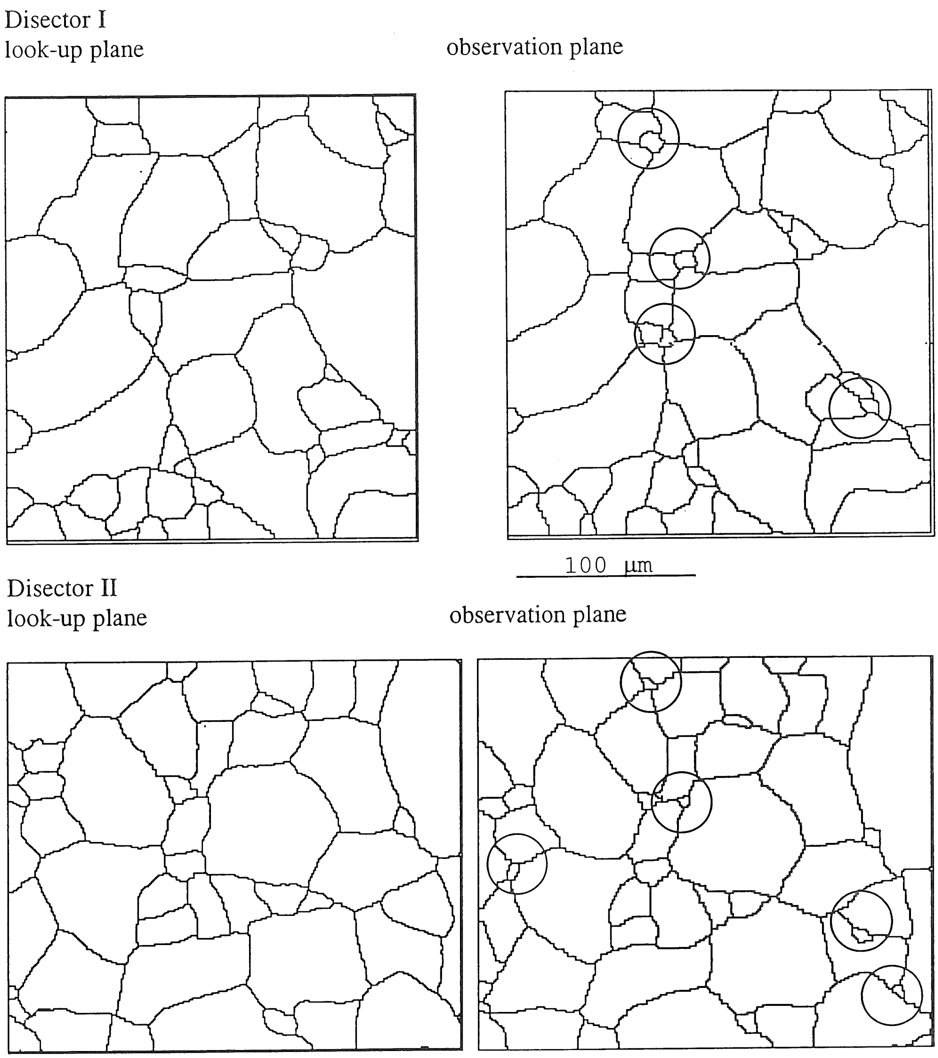

Fig.6. Binary images of grain boundaries in the disector operation.

The preparation of the specimen for the disector method requires a pair of metallographic sections separated by a known distance t. In the present study this parameter varied in the range from 0.8 to 1 μm. Examples of images of grain boundaries before and after removing a layer of material are shown in Fig.6 where "new" grains are circled.

Point sampled intercepts were measured using an image analyzer and specially developed software. Rectangular grids of points were employed and randomly oriented test lines. Length of individual intercepts, l_i, was measured and the mean value of their cubes computed. Measurements of the mean intercept length were carried out on sections parallel to the specimen axis. Such sections meet the criterion of vertical sectioning: e.g. (Baddeley et al. 1986) and accordingly the mean intercept length was computed using a system of test lines.

The mean values and coefficients of variation of the parameters measured directly on 2-D images are given in Table 2.

Table 2. Estimated values of basic parameters characterizing the grain size in the material studied

2D		3D		
$E(A) = 800\ \mu m^2$	$E(d_2) = 28\ \mu m$	$E_N(V) = 40{,}000 \mu m^3$ $E_N(d_3) = 42\ \mu m$	$E_V(V) = 94{,}000\ \mu m^3$ $E_V(d_3) = 56\ \mu m$	$S_V = 0.05\ mm^{-1}$
$CV(A) = 1.0$	$CV(d_2) = 0.5$	$CV(V) = 1.16$		$E(l) = 40\ \mu m$

The measurements carried out using the disector method N_V=2,5*10^{-5} μm^{-3}. This implies that the mean grain volume, $E_N(V)$, equals 40,000 μm^3, ($E_N(V)=1/N_V$), and the mean value of the 3-D equivalent sphere diameter d_3 = 42 μm.

The measurements based on point sampled intercepts resulted in the following estimate of the volume weighted mean volume, $E_V(V)$, $E_V(V)$=94000 μm^3. Estimated values of $E_N(V)$ and $E_V(V)$ yield the volume coefficient of variation, CV(V), equal to 1.16. Such a value of CV(V) confirms a significant spread in the volume of individual grains in polycrystals.

The measurements of the surface area of the grain boundaries in unit volume, S_V, yielded the following estimate of this parameter: $S_V = 0.05\ mm^{-1}$. This implies the mean intercept length, E(l), equal to 40 μm.

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