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Analysis of Spherical Particles Size Distribution – Theoretical Basis

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Abstract

The determination of the form of a probability density function (PDF$_3$) of diameters for nodular particles by using a probability density function (PDF$_2$), which form is empirically estimated from cross-sections of these nodules in a metallographic specimen, can be regarded as a special case of Wicksell's corpuscle problem (WCP). The estimation of the PDF$_3$ for the nodular particles provides information about the kinetics of these particles nucleation, and so about the kinetics of their growth. This information is essential for building more accurate mathematical models of the alloy crystallization.

In the paper there are presented two derivations of the methods used for the estimation of the PDF$_3$ form. The first method bases on diameters received from a planar cross-section. The second one uses also data from the planar cross-section but not the diameters only chords. Both methods provide practical rules for the analysis of the empirical diameters' and chord's size distribution and allow to estimate the mean value of the external surface area of the particles.

Keywords: Estimation of a diameter size distribution, Planimetric analysis, Linear analysis

List of variables used in the text

$C1$ – subclass of $C2$ including only these cut nodules with the radius $r_2 \geq t$ which centres lie closer than $\sqrt{r_3^2 - t^2}$ on both sites of the cutting plane

$C2$ – subclass of $C3$ including only these cut nodules the distance of which from their centre and the cutting plane on both sites is less than r_3

$C3$ – class of nodules with the radii ranging from r_3 to $r_3 + \mathrm{d}r_3$

$\mathrm{d}n_{C1}$ – the cardinality of the set $C1$

$\mathrm{d}n_{C2}$ – the cardinality of the set $C2$

$\mathrm{d}n_{C3}$ – the cardinality of the set $C3$

$E[r_3]$ – the expected value of nodule radii

$E[S_3]$ – the expected value of the outer nodule surface areas

$F_1(t)$ – cumulative distribution function (CDF$_1$) of random chords with the length $2 \cdot r_1 \leq 2 \cdot t$ lying on the cutting plane

$F_2(t)$ – cumulative distribution function (CDF$_2$) of random intersection radii with the length $r_2 \leq t$ lying on the cutting plane

$F_3(t)$ – cumulative distribution function (CDF$_3$) of nodule radii with the length $r_3 \leq t$ in the specimen volume

$f_1(t)$ – probability density function (PDF$_1$) of random chords with the length $2 \cdot r_1 \leq 2 \cdot t$ lying on the cutting plane

$f_2(t)$ – probability distribution function (PDF$_2$) of random intersection radii with the length $r_2 \leq t$ lying on the cutting plane

$f_3(t)$ – probability distribution function (PDF$_3$) of nodule radii with the length $r_3 \leq t$ in the specimen volume

h_1 – distance from a nodule centre to a random chord

h_2 –distance from a nodule centre to a random cutting plane

N_2 – the total number of cut nodules with the cross-section radius $r_2 \geq t$

N_3 –volumetric grain density (grains per unit volume)

r_1 –half-length of a random chord

r_2 –radius of a random cross-section of a nodule

r_3 –radius of a random nodule

R_{max}–the largest radius of a nodule in the probe

$\overline{S}$ – the mean external surface area (estimated value) of nodular particles with different sizes

t –length parameter

x –integration variable

1. Introduction and model assumption

The problem of the mapping of an unknown probability distribution for the size of spherical particles in a non-transparent substance, is one of the classical stereological tasks. The first known solution of this task was proposed by Wicksell [1]. In this paper, a new approach towards this task, for a practical solution, will be presented. In order to derive mathematical formulas, the following assumptions have been used:

- In the specimen volume (Fig. 1) spherical particles with different diameters are redistributed having the volumetric grain density of N_3 (grains per unit volume).
- Spatial distribution of the grain centers is ruled by the Poisson statistical distribution.
- The probability of intersection between particles is negligible.
- The radii distribution is described by an unknown probability density function (PDF$_3$) or by a cumulative distribution function (CDF$_3$).
- The radius of the biggest particle in the probe does not exceed R_{max} value.

For a random section of the probe, the distance h_2 from the particle center to the cutting plane (see Fig. 2) has a uniform statistical distribution. In the same way, the distance h_1 between a random straight test-line (chord) and the centre of the particles has a uniform random variable.

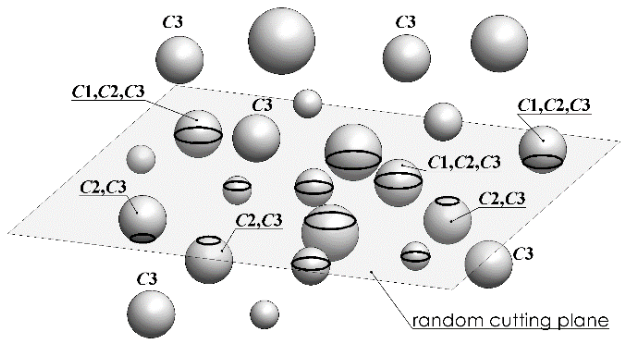

Fig. 1. Particles in the probe space near a cross-section

Let us assume a few things: The number of particles with the radius $r_3 \leq t$ is shown by the equation $F_3(t)$ (the CDF$_3$ of the grain size). The fraction of visible circular sections, lying on the random cutting plane, with a size of $r_2 \leq t$, is described by this function $F_2(t)$ (designated as CDF$_2$). Whereas the fraction of chords placed inside the particles and having the length of $2 \cdot r_1 \leq 2 \cdot t$, is shown by the function $F_1(t)$ designated further as CDF$_1$.

The largest section radius as well as the largest half-length of a chord cannot be bigger than the value of R_{max} of the largest sphere.

Each above-mentioned function $F_i(t)$ corresponds to a probability density function $f_i(t)$ (denoted as PDF$_i$):

$$f_i(t) = \begin{cases} 0 & t < 0 \\ dF_i/dt \quad \text{for} & 0 \leq t < R_{max} \\ 0 & t \geq R_{max} \end{cases} \tag{1}$$

where i = 1 for the half-length of a chord, 2 for the radii of a cross-section, and 3 for the radii of a nodule.

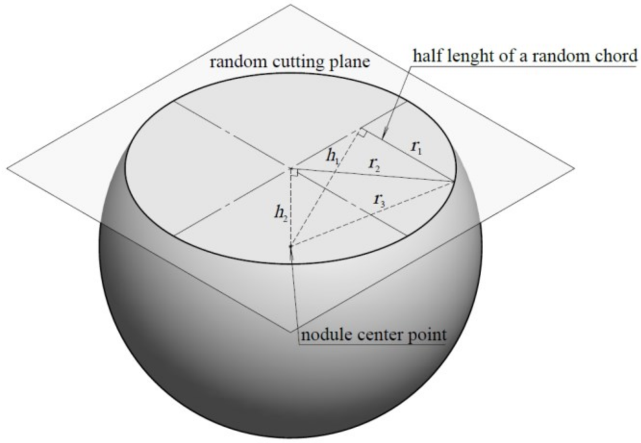

Fig. 2. Illustration of a nodule intersected by a random plane. Legend: r_1 – half-length of a random chord; r_2 – radius of a nodule intersection; r_3 – a nodule radius; h_1 – distance from a nodule center to a chord; h_2 – distance from a nodule center to a random plane

2. Planimetric analysis

Between the nodule radius r_3 and the radius of the section r_2, there is a relation $r_3^2 = r_2^2 + h_2^2$. Getting a section with the radius $r_2 \geq t$ on a nodule with r_3 radius demands the fulfillment of two conditions:

$$r_3 \geq t \tag{2}$$

$$h_2 \leq \sqrt{r_3^2 - t^2} \tag{3}$$

For the uniform spatial distribution of the particles, the probability of meeting the relation (3) is equal to:

$$P\left(h_2 \le \sqrt{r_3^2 - t^2}\right) = \frac{\sqrt{r_3^2 - t^2}}{r_3} \tag{4}$$

In the unit volume of the sample space, let us create a certain class of nodules with radii ranging from r_3 to $r_3 + \mathrm{d}r_3$. Let us denote this class by $C3$. The cardinality $\mathrm{d}n_{C3}$ of this set can be estimated by using the PDF$_3$:

$$\mathrm{d}\,n_{C3}(r_3) = N_3 f_3(r_3)\,\mathrm{d}\,r_3 \tag{5}$$

If in this sample space the random cutting plane is placed, it cuts only those nodules, among the class $C3$, the distance of which from their centre and the cutting plane on both sites is less than r_3. This new subset will be named as $C2$. The cardinality $\mathrm{d}n_{C2}$ per unit area of the class $C2$ can be calculated as follows:

$$\mathrm{d}\,n_{C2}(r_3) = 2 r_3 N_3 f_3(r_3)\,\mathrm{d}\,r_3 \tag{6}$$

Among the class $C2$ the section radii $r_2 \ge t$ will have only the particle which centers are placed at the distance less than $\sqrt{r_3^2 - t^2}$ on the both side of the cutting plane. These particles will establish a new class $C1$ and its cardinality $\mathrm{d}n_{C1}$ can be calculated by multiplication of the Eq. (6) by probability (4):

$$\mathrm{d}\,n_{C1}(t, r_3) = 2\sqrt{r_3^2 - t^2}\, N_3 f_3(r_3)\,\mathrm{d}\,r_3 \tag{7}$$

The total number $N_2(t)$ of the sections with the radius $r_2 \ge t$ can be obtained by integration of the above equation over nodules radius at the range from t to R_{max}:

$$N_2(t) = 2N_3 \int_t^{R_{\max}} f_3(x)\sqrt{x^2 - t^2}\,\mathrm{d}\,x \tag{8}$$

Whereas the number N_2, per unit area of all nodules which are visible on this section can be determined by the integration of Eq. (6) at the range from 0 to R_{max} (or by integration of Eq. (8) from 0 to R_{max}):

$$N_2 = 2N_3 \int_0^{R_{\max}} f_3(x)\, x\,\mathrm{d}\,x \tag{9}$$

As it follows from the above equation, the share of the visible sections with the radius $r_2 \ge t$ on the random plane should be equal to:

$$1 - F_2(t) = \frac{\int_t^{R_{\max}} f_3(x)\sqrt{x^2 - t^2}\,\mathrm{d}\,x}{\int_0^{R_{\max}} f_3(x)\, x\,\mathrm{d}\,x} \tag{10}$$

The integral in the denominator in the Eq. (10) for any statistical distribution function gives the expected (mean) value $E[r_3]$ of the random variable r_3. For that reason, the CDF$_2$ has the following form:

$$F_2(t) = \frac{E[r_3] - \int_t^{R_{\max}} f_3(x)\sqrt{x^2 - t^2}\,\mathrm{d}\,x}{E[r_3]} \tag{11}$$

According to this relation (1) the equation for PDF$_2$ can be obtained by differentiation of the Eq. (11): For the calculation of the integral in the numerator of the above formula, the Leibnitz integral rule was used. According to this rule, the derivative of the integral is equal to:

$$\begin{aligned} &\frac{\mathrm{d}}{\mathrm{d}\,t}\left(\int_t^{R_{\max}} f_3(x)\sqrt{x^2 - t^2}\,\mathrm{d}\,x\right) = \\ &= -\int_t^{R_{\max}} f_3(x)\frac{t}{\sqrt{x^2 - t^2}}\,\mathrm{d}\,x \end{aligned} \tag{12}$$

With regard to (12) the form of PDF$_2$ is:

$$f_2(t) = \frac{t}{E[r_3]}\int_t^{R_{\max}} \frac{f_3(x)}{\sqrt{x^2 - t^2}}\,\mathrm{d}\,x \tag{13}$$

Having the form of $f_2(t)$, which is estimated by quantitative metallography results, the equation (13) can be solved implicitly with respect to the $f_3(x)$ function. In this task such a solution (named sometimes as inverse) according to [2] gives unsatisfactory results. That is why the numerical solutions of this task are used most often. An analysis of the planar section is based directly on Wickesell equation (13) which has been used for volume size distribution of spheroidal particles by Scheil [3], Schwartz [4], Saltykov [5, 6], Li at al. [7]. A similar solution for the mineralogy task has been presented in [8]. Unfortunately, small numerous errors of the empirically estimated function $f_2(t)$ result in the *„arbitrarily large perturbations of the solution"* [9, 10]. In this context, Eq. (13) is used usually not for designing the PDF$_3$ form, but for the examination of matching the empirical function and one of the selected statistical distribution laws, e.g. normal, log-normal, Weibull or uniform-sized [2, 7, 11].

3. Linear analysis

The fallowing relation $r_3^2 = r_1^2 + h_1^2$ links the nodular particle radius r_3 and half-length of the random chord r_1 on the random plane, where h_1 is the distance from the particle center to the chord line (see Fig. 2). The nodules of radius r_3 will have a chord length $2r_1 \geq 2t$ under the following conditions:

$$r_3 \geq t \tag{14}$$

$$h_1 \leq \sqrt{r_3^2 - t^2} \tag{15}$$

For uniform spatial distribution of particles, the probability of relation (15) is equal to:

$$P\left(h_1 \leq \sqrt{r_3^2 - t^2}\right) = \frac{r_3^2 - t^2}{r_3^2} \tag{16}$$

Cardinality of $C3$ class particles is estimated by Eq. (5). The section number of C3 class particles by a random straight line per unit length of this line should be estimated as follows:

$$\mathrm{d}\, n_1(r_3) = \pi r_3^2 N_3 f_3(r_3) \mathrm{d}\, r_3 \tag{17}$$

The number of chords longer than $2t$ for $C3$ particles can be calculated by the multiplication of the previous equation by probability (16):

$$\mathrm{d}\, n_1(t, r_3) = \pi \left(r_3^2 - t^2\right) N_3 f_3(r_3)\, \mathrm{d}\, r_3 \tag{18}$$

The total amount $N_1(t)$ of the bisecant longer than $2t$ should be obtained by the integration of the above equation over nodules radius at the range from t to R_{max}:

$$N_1(t) = \pi N_3 \int_t^{R_{max}} \left(x^2 - t^2\right) f_3(x)\, \mathrm{d}x \tag{19}$$

The total amount of all visible bisecants at the random plane section should be determined by the integration of Eq. (19) with an inferior limit of integration equal to 0 (or by integration of Eq. (17) from 0 to R_{max}):

$$N_1 = \pi N_3 \int_0^{R_{max}} x^2 f_3(x) \mathrm{d}\, x \tag{20}$$

As it follows from the above formula, a fraction of the chords longer than $2t$, among all chords which are visible on the nodular particles on the random plane, should be equal to:

$$1 - F_1(t) = \frac{\int_t^{R_{max}} \left(x^2 - t^2\right) f_3(x) \mathrm{d}\, x}{\int_0^{R_{max}} x^2 f_3(x) \mathrm{d}\, x} \tag{21}$$

The mean external surface (estimated value) of the nodular particles of different sizes is equal to:

$$\bar{S} = E[S_3] = 4\pi \int_0^{R_{max}} f_3(x) x^2 \mathrm{d}\, x \tag{22}$$

Thence, relation (21) should be transformed to form:

$$F_1(t) = \frac{\bar{S} - 4\pi \int_t^{R_{max}} f_3(x)\left(x^2 - t^2\right) \mathrm{d}x}{\bar{S}} \tag{23}$$

According to relation (1) the equation for PDF$_1$ should be obtained by differentiation of the Eq. (23). According to Leibnitz integral rule, the derivative of the integral in Eq. (23) is equal to:

$$\frac{\mathrm{d}}{\mathrm{d}\, t}\left(\int_t^{R_{max}} f_3(x)\left(x^2 - t^2\right) \mathrm{d}x\right) = -2t \int_t^{R_{max}} f_3(x) \mathrm{d}\, x \tag{24}$$

Taking into account the derivative (24), the following form of the equation for PDF$_1$ should be proposed as:

$$f_1(t) = \frac{8\pi t}{\bar{S}} \int_t^{R_{max}} f_3(x) \mathrm{d}\, x \tag{25}$$

According to the PDF$_3$ definition, the integral equation (25) may be transformed to form:

$$f_1(t) = \frac{8\pi t}{\bar{S}} \left[F(R_{max}) - F_3(t)\right] \tag{26}$$

Because $F_3(R_{max}) = 1$:

$$f_1(t) = \left[1 - F_3(t)\right] \frac{8\pi t}{\bar{S}} \tag{27}$$

The above means that the empirical CDF$_3$ should be estimated on the basis of the empirical estimation of PDF$_1$ as follows:

$$F_3(t) = 1 - \frac{\bar{S}}{8\pi t} f_1(t) \tag{28}$$

The derivations of Eq. (28) with respect t parameter gives:

$$f_3(t) = \frac{\bar{S}}{8\pi}\left[\frac{f_1(t)}{t^2} - \frac{1}{t}\frac{\mathrm{d}\, f_1(t)}{\mathrm{d}\, t}\right] \tag{29}$$

This formulation corresponds to known solutions of Cahn and Fullmann [12], Lord and Willis [13], and Spektor [14]:

$$N_3 f_3(t) = \frac{N_1}{2\pi}\left[\frac{f_1(t)}{t^2} - \frac{1}{t}\frac{\mathrm{d}\, f_1(t)}{\mathrm{d}\, t}\right] \tag{30}$$

4. Practical rules of the analysis

As it follows from Eq. (27), for $F_3(t) < 1$ (or $t < R_{max}$) at the intervals ($t_1 \ldots t_2$), when condition $\mathrm{d}F_3(t)/\mathrm{d}t = 0$ is fulfilled, value of PDF_1 will be proportional to the t parameter. This condition is true when there are no particles of radius $t_1 \leq t < t_2$ in the space of the probe. In such a situation, the proportionality factor will depend on the particle's share of radius less than t_1 and on the mean external surface of the nodules:

$$k(t) = \frac{f_1(t)}{t} = [1 - F_3(t_1)]\frac{8\pi}{\bar{S}} \tag{31}$$

For $t_1 = 0$ and a good resolution of image analysis (this means that optical resolution is much better than t_2) it is possible to estimate the value of the mean external surface of nodules as follows:

$$\bar{S} = \frac{8\pi t_2}{f_1(t_2)} \tag{32}$$

As it follows from Eq. (27), regardless of the nature of the CDF_3 $\lim_{t \to R} f_1(t) = 0$.

Accuracy of $\bar{S}$ estimation by means of Eq. (30) should be verified by the integration of:

$$\bar{S} = 4\pi \int_0^{R_{max}} t^2 f_3(t)\,\mathrm{d}\, t \tag{33}$$

5. Conclusions

- Empirical form of CDF_3 of the sizes of nodular particles should be estimated on the basis of the empirical measurement of PDF_1 on the statistical distribution of the length of random chords by Eq. (28).
- If in the tested probe there are no nodular particles of radii $t_1 \leq t < t_2 < R_{max}$, then at the range between t_1 and t_2 the PDF_3 value is proportional to t.
- If in the space of the analyzed probe there are no spheroids of radii less than t_2, then in the range between 0 and t_2 the PDF_1 value is proportional to t. The proportionality factor is equal to $8\pi/\bar{S}$, where $\bar{S}$ is the mean value of the external surface of the nodules.

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