DEPENDENCE OF CHEMICAL INHOMOGENEITY OF GRANULAR MATERIALS ON THE SIZE AND SHAPE OF PARTICLES

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Avenues have been explored of possible improvement of inhomogeneity of a granular material using the relationships defining the structure of chemical inhomogeneity of compact and granular materials and a proper choice of particle size, shape and orientation in the original compact material. The results apply generally to granular materials prepared from compact (monolitic) samples. It is apparent though that considerations regarding optimum shape and orientation of the particles concern mostly metallic materials in the form of cutting chips while do not pertain to powdered materials prepared by grinding. The used method consisted of the assessment of the dependence of individual components of inhomogeneity on particle size for various shapes and orientations. There were essentially three types of dependences found: Non-periodic components of inhomogeneity of the granular material depend on particle size and/or shape only slightly; periodic components diminish starting from a certain value with growing volume of the particle. The regression component increases with the particle's volume. The contradicting character of the two latter types of dependences pointed at the possibility of existence of a minimum (apart from the trivial case of $V = 0$) on the course of the dependence of the overall variance of inhomogeneity of the granular material *versus* particle volume. This minimum corresponds to the sought optimum particle volume. Position of this minimum and hence the optimum volume of the particle depend generally on the shape of the particle and its orientation within the material.

Relatively voluminous literature has been devoted to the problem of inhomogeneity of granular materials. Its review can be found ref.[1]. In this text we shall examine another aspect of this problem, namely the quantitative relationship between chemical inhomogeneity of a granular material and particle size and shape. Quantitatively, the inhomogeneity of materials is usually characterized by the inhomogeneity variance, *i.e.* by one of the components of the overall variance of results of chemical analyses. The other component, as it is well known, is the variance of the measuring method.

The inhomogeneity variance appears due to concentration fluctuations of the analyzed elements in the material, and, in dependence on the analytical method used, on the nonuniform distribution of other elements affecting the analysis. Various defects of crystal lattice may eventually contribute too.

More conspicuously the variance of inhomogeneity shows particularly on analyses of small samples encountered in modern analytical methods. This calls for the preparation of as homogeneous materials as possible to serve as analytical standards.

Inhomogeneity of granular materials may be diminished by improved homogeneity of the compact (monolitic) material used for preparation, by mixing (the question of mixing has been investigated in a number of papers summarized in ref.[1]) and by the choice of the size and shape of the particles prepared from the original sample. The problems associated with the latter mechanism shall be dealt with in the following text. The approach shall utilize results of our earlier studies devoted to the determination of inhomogeneity of compact materials[2] and the determination of inhomogeneity of granular materials[3] from the knowledge of the characteristics of the inhomogeneity of the compact material used for the preparation of the granular sample. It is assumed that the basic data regarding the distribution of the analyzed element in the compact material are obtained by the electron microprobe. The volume of the sample for the microprobe, *i.e.* the part of the material analyzed in a single analysis[4], is in the order of magnitude 10 μm^3. The approaches presented in ref.[2] enable the inhomogeneity variance to be computed from these data for larger samples of an arbitrary shape, *i.e.* also for samples and shapes of particles prepared from the original material. The variance of inhomogeneity of samples-particles – (one-particle samples) – represent in the modification of ref.[3] a principal datum for the calculation of inhomogeneity of samples consisting of several such particles such as those analyzed by classic analytical methods.

Principal Concepts and Relations

The course of concentration c of an analyzed element in a compact material may be found on various cuts through the material by the electron microprobe (see *e.g.* the paper[5] devoted to the determination of inhomogeneity of silumine). The measured concentrations may in turn serve to provide estimates of the mean concentration $\bar{c}$. In addition, one can obtain by regression the course of local mean concentrations c_r, which are generally different from $\bar{c}$.

If the material consists of grains of m phases (designated by the index $i = 1, 2, \ldots, m$), we can also measure the phase concentrations c_i (*i.e.* concentrations of the analyzed element in the i-th phase) and determine their overall means $\bar{c}_i$ as well as the regression estimate of the course of local means c_{ir}. The data of the microprobe may further serve (ref.[5]) to provide corresponding estimates of the volume concentration of the i-th phase v_i, v_{ir} and the densities s_i, s_{ir}.

The above introduced quantities shall be used to express the deviation of $\bar{c}$ for the concentration $c(\omega)$ measured in a sample ω

$$c(\omega) - \bar{c} = [c(\omega) - c_{hr}(\omega)] + [c_{hr}(\omega) - c_r] + [c_r - \bar{c}] . \qquad (1)$$

The quantity $c_{hr}(\omega)$ has been introduced to characterize the inhomogeneity of the material. It designates the concentration prevailing in the sample ω, had the phase concentrations c_i within each phase not been fluctuating and had they equaled their local (regressed) mean values c_{ir}. For $c_{hr}(\omega)$ we thus may write

$$c_{hr}(\omega) = \sum_{i=1}^{m} c_{ir} v_i(\omega)\, s_{ir}/s_{hr}(\omega) \tag{2}$$

where $v_i(\omega)$ is the true volume concentration of the i-th phase in ω and $s_{hr}(\omega) = \sum_{i=1}^{m} v_i(\omega)\, s_{ir}$ stands for the density of the sample ω under the assumption $c_i \equiv c_{ir}$.

The deviations $[c(\omega) - \bar{c}]$ in Eq. (*1*) have their corresponding inhomogeneity variance $D_J^2(\omega)$ belonging to the set of samples ω. The deviations $[c(\omega) - c_{hr}(\omega)]$ occur due to concentration fluctuations within the phases and the corresponding variance is termed the intraphase variance designated by $D_f^2(\omega)$. The deviations $[c_{hr}(\omega) - c_r]$ show through even presence of grains of different phase in samples ω and their corresponding variance is $D_h^2(\omega)$ while the deviations $[c_r - \bar{c}]$ have corresponding regression variance $D_r^2(\omega)$.

The inhomogeneity variances $D_J^2(\omega)$ and its components $D_f^2(\omega)$, $D_h^2(\omega)$ and $D_r^2(\omega)$ are related (neglecting possible correlation of the deviations[2] $[c(\omega) - c_{hr}(\omega)]$ and $[c_{hr}(\omega) - c_r]$) by

$$D_J^2(\omega) = D_f^2(\omega) + D_h^2(\tau) + D_r^2(\omega)\,. \tag{3}$$

A full characterization of inhomogeneity of a material necessitates the knowledge of the dependence of the components of inhomogeneity on the size and shape of the sample ω. On taking into considerations only the samples of particles of the size of practical importance (particles to be prepared from the original material), the regression variance $D_r^2(\omega)$ may be regarded as independent of the size and shape of the sample ω (and as such may be simply designated by D_r^2). This is so because in the practical range of particle size the course of the concentration c_r within the sample may be taken with sufficient accuracy to be linear. Hence the regression estimate for the whole sample equals c_r in the center of the sample regardless of its shape and size.

In case of the intraphase variance the situation is different. For the determination of $D_f^2(\omega)$ from the deviations $[c(\omega_{f0}) - c_{hr}(\omega_{f0})]$ (ω_{f0} is the sample analyzed by the microprobe) it is necessary to construct the correlation function and to determine from it the periodic $D_{fp}^2(\omega)$ and the nonperiodic part $D_{fn}^2(\omega)$ of the intraphase variance $D_f^2(\omega)$ ($D_f^2(\omega) = D_{fp}^2(\omega) + D_{fn}^2(\omega)$).

In the paper[2] the variance $D_{fp}^2(\omega)$ was computed from a model which shall be also used here as a starting point: It is assumed that the periodic part of the concentration fluctuation is given by $A \cos(2\pi x_1/\alpha_1) \cos(2\pi x_2/\alpha_2) \cos(2\pi x_3/\alpha_3)$, where α_1, α_2, α_3

are the periods in the directions of axis x_1, x_2, x_3 of a rectangular coordinate system fixed to the examined compact material. It is also assumed that the samples ω (in this text identical with the particles to be later prepared from the compact material), have the shape of parallelepipeds and that the edges of these parallelepipeds, b_1, b_2, b_3, are parallel to the axes x_1, x_2, x_3. For the variance $D^2_{fp}(\omega)$ we then have two expressions derived: One accurate, not presented here, and, the other, a simplified one, given by (in order to express the dependence of the variance on the volume $V = b_1 \,.\, b_2 \,.\, b_3$ of the sample ω, or on the given ratio $b_1 : b_2 : b_3$, we shall use the symbols $D^2_{fp}(V)$ or $D^2_{fp}(b_1 \,.\, b_2 \,.\, b_3)$ instead of $D^2_{fp}(\omega)$ and similarly for the remaining components of the variance of inhomogeneity):

$$D^2_{fp}(b_1 \,.\, b_2 \,.\, b_3) \approx D^2_{fpo}(b_1 \,.\, b_2 \,.\, b_3) = A^2 K_{10} K_{20} K_{30} \,. \tag{4}$$

For the quantities $K_{i0} (i = 1, 2, 3)$ we may write: if $b_i \leqq b_{i0} = \alpha_i/n$, then $K_{i0} = C_{fi}$; if $b_i > b_{i0}$, then $K_{i0} = C_{ji}(\alpha_i/\pi)^2 \, b_i^{-2}$. The constant C_{fi} depends on the ratio of the period α_i to the size of the compact material in the direction of the axis x_i. (If α_i is substantially less than the size of the material in the corresponding direction than $C_{fi} \approx 1/2$. If, on the contrary, α_i is subtantially greater than the size of the material in the corresponding direction then $C_{fi} = 1$. In practice this, of course, indicates that there exist no periodicity in the given direction). The amplitude A is determined from measurements by the microprobe, with the aid of the already mentioned correlation function the variance $D^2_f(\omega_{fc})$, measured by the microprobe, is split into the periodic part $D^2_{fp}(\omega_{fo})$ and the nonperiodic part $D^2_{fn}(\omega_{fo})$. For A we then may write: $A^2 = 2D^2_{fp}(\omega_{fo})$.

For the nonperiodic part of the intraphase variance we shall use the approximate formula of the form[2]

$$D^2_{fn}(V) = \begin{cases} D^2_{fn}(\omega_{fo}) & \text{for} \quad V \leqq V_{fn} \\ D^2_{fn}(\omega_{fo}) \, V_{fn}/V & \text{for} \quad V > V_{fn} \,. \end{cases} \tag{5}$$

In the previous paper the volume V was designated by ka^3, where a^3 represented the corrected volume of the sample ω_{fo} analyzed by the microprobe. The constant V_{fn} then must be expressed in the form $k_0 a^3$; the method of determining k_0 has been published earlier[2].

Existing methods of determination[2] of the heterogeneity variance are those for samples smaller than the volume of the grain of the phases, as well as those for samples with at least one dimension substantially greater than the dimensions of the phase-homogeneous region of the material. In the first case, the variance of inhomogeneity is computed from

$$D^2_h(V) = D^2_h(0) - V^{1/3} \lambda \tag{6}$$

where $D_{\mathrm{h}}^2(0)$ designates the variance of heterogeneity of infinitesimaly small samples and λ is a constant depending on the scatter of the phase (the determination of both constant has been described in ref.[2]). The shape of the sample – the particle of volume V – is in accord with the assumptions cubic, which is thought to be a sufficient approximation for powdered materials to which Eq. (*6*) applies.

For the determination of the variance of heterogeneity of samples with at least one dimension substantially larger than the size of the phase-homogeneous regions in the compact material, the method of parallelepipeds has been worked out[2]. According to this method ω may be thought to be composed of parallelepipeds (designated by ω_{h0}), whole lateral dimension is given by the depth of the layer analyzed by the microprobe. Its length must be taken so as to provide that each parallelepiped consist of a larger number of grains of different phases. On a photograph of the surface of the cut through the material we draw a set of parallel parallelepipeds, created by repeated shifting of the first parallelepiped in a chosen direction. For individual parallelepipeds we then determine the concentrations $c_{\mathrm{hr}}(\omega_{\mathrm{h0}})$ and further procedure is analogous to the determination of the intraphase variance: The deviations $[c_{\mathrm{hr}}(\omega_{\mathrm{h0}}) - c_{\mathrm{r}}]$ is used to construct the correlation function for the given direction and its periodic and the nonperiodic parts serve to calculate: *i*) the periodic and the nonperiodic parts of the variance of heterogeneity corresponding to the samples ω_{h0} – the parallelepipeds (*i.e.* $D_{\mathrm{hp}}^2(\omega_{\mathrm{h0}})$ and $D_{\mathrm{hn}}^2(\omega_{\mathrm{h0}})$), and *ii*) additional parameters necessary for the determination of the periodic part $D_{\mathrm{hp}}^2(\omega)$ (or $D_{\mathrm{hp}}^2(V)$, or $D_{\mathrm{hp}}^2(b_1 \cdot b_2 \cdot b_3)$) and the nonperiodic part $D_{\mathrm{hn}}^2(\omega)$ (or $D_{\mathrm{hn}}^2(V)$, $D_{\mathrm{hn}}^2(b_1, b_2, b_3)$) of the heterogeneity variance of the samples ω from Eqs (*4*) and (*5*) in which the subscript f is replaced by the subscript h. On assuming that the parallelepipeds are oriented in the direction of axis x_1, we substitute for K_{10} for Eq. (*4*), for b_{10} the larger of the values L, α_1/π and for K_{20} and K_{30} we substitute $b_{20} = \alpha_2/\pi$ and $b_{30} = \alpha_3/\pi$. There is, however, an alternative way: put $b_{10} = \alpha_1/\pi$ even in the case that $\alpha_1/\pi < L$ and simultaneously express the constant A from Eq. (*4*) in a modified form as $A = D_{\mathrm{hp}}(\omega_{\mathrm{h0}}) \sqrt{2}\, \pi L/\alpha_1$ in order to keep a similar course for $b_1 > L$. This alternative shall be used in the following text for its closer analogy to the intraphase variance.

The constant V_{hn} for Eq. (*5*) is determined according to ref.[2] by calculating the cross sectional area of the characteristic cluster of parallelepipeds S_{hn} (earlier[2] designated $k_0 d$) and put $V_{\mathrm{hn}} = LS_{\mathrm{hn}}$.

After obtaining the parameters necessary for the determination of the components of the inhomogeneity variance (*i.e.* D_{r}^2, $D_{\mathrm{fp}}^2(\omega)$, $D_{\mathrm{fn}}^2(\omega)$ and $D_{\mathrm{h}}^2(\omega)$, or $D_{\mathrm{hp}}^2(\omega)$ and $D_{\mathrm{hn}}^2(\omega)$) of the compact material we can use the methods published in ref.[3] to determine corresponding components of the variance of inhomogeneity of the granular material prepared from the original compact material.

The granular materials shall be bassumed to be perfectly mixed (an exact definition of this state including references has been published elsewhere[3]). For such materials the dependence of inhomogeneity on the size of the sample shall be expressed by the familiar formula

$$D_J^2(k\omega) = k^{-1} D_J^2(\omega)\,, \tag{7}$$

where $k\omega$ designates the sample k-times greater by weight than the sample ω.

Thus on dividing the compact material into particles of the same size and shape the knowledge sufficient for the overall characterization of inhomogeneity of the prepared granular material is that of a single parameter – the variance of inhimogeneity of the corresponding one-particle samples. From the knowledge of the parameters of inhomogeneity of the compact material we are able such a variance of sample of later prepared particles identical as to size and shape to determine[2,3].

In the following text we shall confine ourselves to the already somewhat simplified model according to which all particles of the granular material have the shape of a $b_1 . b_2 . b_3$ parallelepiped. Corresponding one-particle variance is designated $D^2(b_1 . b_2 . b_3)$. For a set of 1 gram samples for example we obtain according to Eq. (7) the variance of inhomogeneity in the form $D^2(b_1 . b_2 . b_3)/\bar{k}$ where $\bar{k}$ is the mean number of particles in a single sample (the weights of the particles are not generally the same). On designating by v_0 the volume of a 1 gram sample, then $\bar{k} = v_0/(b_1 . b_2 . b_3)$. By preparing from the same compact material particles of different size or shape (other ratio $b_1 : b_2 : b_3$) we obtain for the given set of 1 gram samples a generally different variance for we have a different particle variance as well as a different mean number of particles in the set.

The variance of inhomogeneity of the set of 1 gram samples, which shall be designated by $D^2(1\text{ g}; b_1 . b_2 . b_3)$, may be taken for the criterion of suitability of the choice of the size and shape of the particles. As already mentioned, Eq. (7) gives the following transformation

$$D^2(1\text{ g}; b_1 . b_2 . b_3) = (b_1 . b_2 . b_3/v_0)\, D^2(b_1 . b_2 . b_3)\,. \tag{8}$$

Using this relation one can transform individual components of the inhomogeneity variance in the compact material and their dependences on the size and shape of the sample into corresponding dependences of the inhomogeneity of the granular material represented by the variance $D^2(1\text{ g}; b_1 . b_2 . b_3)$.

The Dependence of Inhomogeneity of the Granular Material on the Particle Size

In this paragraph we shall compare sets of particles prepared from the same compact material while it is assumed that individual sets differ only in the volume of the particles and their shape (the ratio $b_1 : b_2 : b_3$ is the same for all sets).

Eq. (*8*) may then be replaced by a simpler equation

$$D^2(1\ \text{g};\ V) = (V/v_0)\, D^2(V)\,, \tag{9}$$

where $V = b_1 \,.\, b_2 \,.\, b_3$ is the volume of the particle of the examined granular material.

As above mentioned the regression component of the one-particle variance is independent of particle volume: $D_r^2(V) = D_r^2 = \text{const}$. Substituting into Eq. (*9*) we arrive at corresponding dependence of the component $D_r^2(1\ \text{g};\ V)$ on V

$$D_r^2(1\ \text{g};\ V) = (V/v_0)\, D_r^2\,. \tag{10}$$

The regression component of the variance of 1 g samples thus grows linearly with particle volume. The plot is a straight line passing through the origin (Fig. 1, curve r)

For calculation of the nonperiodic component of the intraparticle variance and the variance of heterogeneity, we have the same formula (*5*). Both components shall be therefore solved for simultaneously omitting the index f or h and retaining only the index n. Substituting Eq. (*5*) into (*9*) we obtain

$$D_n^2(1\ \text{g};\ V) = \begin{cases} (V/v_0)\, D_n^2(\omega_0) & \text{for} \quad V \leqq V_n \\ (V_n/v_0)\, D_n^2(\omega_0) & \text{for} \quad V > V_n \end{cases} \tag{11}$$

A graphical form of Eq. (*11*) is the curve n in Fig. 1: for $V \leqq V_n$ the variance $D_n^2(1\ \text{g};\ V)$ grows with the volume V linearly from zero up to $(V_n/v_0)\, D_n^2(\omega_0)$; for $V \geqq V_n$ it remains constant. The periodic components of the intraphase variance of heterogeneity, for which Eq. (*4*) applies, shall be solved for also simultaneously. Formula (*4*), however, shall be first rearranged into a form more suitable for the study of the variance of heterogeneity D_p^2 as a function of particle volume V. (The derivation of a general and more accurate dependence of the variance $D_p^2(1\ \text{g};\ V)$ on the volume V shall be described below). Assuming that the periods $\alpha_1, \alpha_2, \alpha_3$ are finite, the variance $D_p^2(V)$ may be expressed in the form analogous to Eq. (*5*)

$$D_p^2(V) = \begin{cases} D_p^2(\omega_0)\, C & \text{for} \quad V \leqq V_p \\ D_p^2(\omega_0)\, C V_p^2/V^2 & \text{for} \quad V > V_p\,. \end{cases} \tag{12}$$

The constant C is given by $C = C_0 \,.\, C_1 \,.\, C_2 \,.\, C_3$. The constants $C_1 \,.\, C_2 \,.\, C_3$ appear implicitly in Eq. (*4*); the constant C designates the coeffient of $D_p^2(\omega_0)$ in the expression for the amplitude of the oscillations squared A^2(*i.e.* for the intraphase variance $C_0 = 2$ and for the variance of heterogeneity either $C_0 = 2$ or $C = 2(\pi L/\alpha_1)^2$. The characteristic volume V_p is defined by $V_p = b_{10} \,.\, b_{20} \,.\, b_{30} = \alpha_1 \,.\, \alpha_2 \,.\, \alpha_3 \,.\, \pi^{-3}$.

Substituting Eq. (*12*) into (*9*) for $D^2(1\text{ g};V)$ we obtain

$$D_{\mathrm{p}}^2(1\text{ g};V)=\begin{cases}(V/v_0)\,D_{\mathrm{p}}^2(\omega_0)\,C & \text{for}\quad V\leqq V_{\mathrm{p}}\\ (V_{\mathrm{p}}/v_0)\,D_{\mathrm{p}}^2(\omega_0)\,CV_{\mathrm{p}}/V & \text{for}\quad V>V_{\mathrm{p}}\,.\end{cases}\tag{13}$$

Corresponding curve is the curve *p* in Fig. 1: in its first part ($V\leqq V_{\mathrm{p}}$) the variance $D^2(1\text{ g};V)$ grows linearly from 0 to $(V_{\mathrm{p}}/v_0)\,D_{\mathrm{p}}^2(\omega_0)\,C$; in the second part ($V\geqq V_{\mathrm{p}}$) it decreases with the first power.

For the interval of validity of Eq. (*6*) we obtain (by substituting (*6*) into Eq. (*9*)) for the variance of heterogeneity of 1 g samples the following expression

$$D_{\mathrm{h}}^2(1\text{ g};V)=(V/v_0)\,D_{\mathrm{h}}^2(0)-(V^{4/3}/v_0)\lambda\,.\tag{14}$$

The function (*14*) may be approximated by the curve *n* in Fig. 1: the variance $D(1\text{ g};V)$ (or its estimate $D^2(1\text{ g};V)$) grows linearly with V from zero up to $V_{\mathrm{h}}=0{\cdot}1D_{\mathrm{h}}^6(0)\,\lambda^{-3}$ where it reaches $0{\cdot}1D_{\mathrm{h}}^8(0)\,\lambda^{-3}$. Its further course is a constant while for volumes V approaching the size of the phase homogeneous regions in the original compact material Eqs (*6*) and (*14*) are no longer valid.

The course of the overall variance of inhomogeneity $D_{\mathrm{i}}^2(1\text{ g};V)$ of 1 g samples of the granular material in dependence on the volume V of particles of constant shape may thus be represented by the curve *i* which is a combination of the curve *r* and several curves *p* and *n*. Fig. 1 is only a simplified representation of a probably more complicated reality (there exist probably more curves of type *p* and *n*). Nevertheless, it clearly evidences the possibility of existence of a minimum variance of inhomogeneity variance of the granular material $D_{\mathrm{i}}^2(1\text{ g};V_0)$ at an optimum volume of particles (not considering of course the trivial case $V=0$). A condition for the existence of the minimum is that the curve *p* have at the point $V=V_{\mathrm{p}}$ a value larger than that

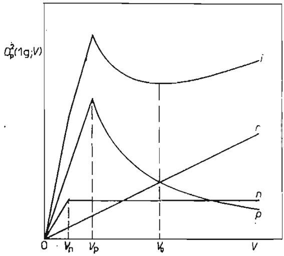

FIG. 1

Dependence of the Inhomogeneity Variance (*i*) and its Components (*r*, *p*, *n*) for a Set of 1 gram Samples on Volume V of a Particle of a Granular Material Consisting of Identical Particles on the curve r. By comparing Eqs (*10*) and (*13*) we obtain for this stimulation the expression

$$D_p^2(\omega_0)\, C > D_r^2 . \qquad (15)$$

Choice of Particle Shape and Their Orientation within the Material

It has been noted that the regression variance D_r^2 of one-particle samples is independent of both the size and shape of the particles. Eq. (*10*) then gives that the regression variance of 1 g samples $D_r^2(1\text{ g};\, b_1 . b_2 . b_3)$ depends only on the volume $V = b_1 . b_2 . b_3$ of the particles and not on their shape, *i.e.* on the ratio $b_1 : b_2 : b_3$.

For the periodic part of the intraphase variance or the heterogeneity variance the situation is more complicated. In order to examine this case we shall construct the dependences of $D_p^2(1\text{ g};\, b_1 . b_2 . b_3)$ on the volume $V = b_1 . b_2 . b_3$ for various shapes of particles, *i.e.* for various ratios $b_1 : b_2 : b_3$ (for a given graph the ratio $b_1 : b_2 : b_3$ is constant).

It is assumed again that the periodic component of the concentration fluctuation within the compact material has in the direction of the axes x_1, x_2, x_3 of a rectangular system firmly fixed to the compact material periods $\alpha_1, \alpha_2, \alpha_3$ and that the particles shaped as parallelepipeds of size b_1, b_2, b_3 were in its original position oriented within the compact material axially (*i.e.* $b_i \parallel x_i$, $i = 1, 2, 3$) and, further, that the shape of the particle is the same and fixed by the ratio $b_1 : b_2 : b_3$.

The dependence of the one-particle sample variance $D_p^2(b_1, b_2, b_3)$ on the volume $V = b_1, b_2, b_3$ shall be computed from Eq. (*4*). Proceeding from $V = 0$ the character of the dependence in Eq. (*4*) varies whenever some of the edges b_i reaches α_i/π in size. Suppose that this critical value is reached first by the edge b_3 and designate in this case its size by b_{31} $(b_{31} = \alpha_3/\pi)$ the size of the remaining edges is b_{11} and b_{21} and corresponding volume $V_1 = b_{11} . b_{21} . b_{31}$. With further increase of the volume let the characteristic size be reached by the edge b_1. Analogously we designate b_{12} $(b_{12} = \alpha_1/\pi)$ and $V_2 = b_{12} . b_{22} . b_{32}$. Finally, the critical size is reached by the edge b_2 and we designate corresponding value by b_{23} $(b_{23} = \alpha_2/\pi)$ and corresponding volume by $V_3 = b_{13} . b_{23} . b_{33}$.

In individual intervals the one-particle variance $D_p^2(b_1 . b_2 . b_3)$ computed from Eq. (*4*) and corresponding variance of one-gram samples computed from Eq. (*9*) is $D_p^2(1\text{ g};\, b_1 . b_2 . b_3)$ are given by:

The first interval $0 < V \leqq V_1$, $(0 < b_1 \leqq b_{11}, 0 < b_2 \leqq b_{21}, 0 < b_3 \leqq b_{31})$

$$D_p^2(b_1 . b_2 . b_3) = D_p^2(\omega_0)\, C \qquad (16)$$

$$D_p^2(1\text{ g};\, b_1 . b_2 . b_3) = (1/v_0)\, D_p^2(\omega_0)\, C\, b_1 . b_2 . b_3 . \qquad (17)$$

The second interval $V_1 \leqq V \leqq V_2$, $(b_{i1} \leqq b_i \leqq b_{i2})$

$$D_p^2(b_1 \,.\, b_2 \,.\, b_3) = D_p^2(\omega_0)\, C(b_{31}/b_3)^2 \qquad (18)$$

$$D_p^2(1\ \mathrm{g};\, b_1 \,.\, b_2 \,.\, b_3) = (1/v_0)\, D_p^2(\omega_0)\, C(\alpha_3/\pi)^2 \,.\, b_1 \,.\, b_2/b_3 \,. \qquad (19)$$

The third interval $V_2 \leqq V \leqq V_3$, $(b_{i2} \leqq b_i \leqq b_{i3})$

$$D_p^2(b_1 \,.\, b_2 \,.\, b_3) = D_p^2(\omega_0)\, C(b_{31}/b_3)^2\, (b_{12}/b_1)^2 \qquad (20)$$

$$D_p^2(1\ \mathrm{g};\, b_1 \,.\, b_2 \,.\, b_3) = (1/v_0)\, D_p^2(\omega_0)\, C(\alpha_3/\pi)^2\, (\alpha_1/\pi)^2\, b_2(b_3 b_1)^{-1} \,. \qquad (21)$$

The fourth interval $V > V_3 (b_i > b_{i3})$

$$D_p^2(b_1 \,.\, b_2 \,.\, b_3) = D_p^2(\omega_0)\, C(b_{31}/b_3)^2\, (b_{12}/b_1)^2\, (b_{23}/b_2)^2 \qquad (22)$$

$$D_p^2(1\ \mathrm{g};\, b_1 \,.\, b_2 \,.\, b_3) = (1/v_0)\, D_p^2(\omega_0)\, C(\alpha_1 \,.\, \alpha_2 \,.\, \alpha_3)^2\, \pi^{-6} \,.\, (b_1 \,.\, b_2 \,.\, b_3)^{-1} \,. \qquad (23)$$

The course of the dependence of the variance $D_p^2(1\ \mathrm{g};\, b_1 \,.\, b_2 \,.\, b_3)$ on particle volume V in the first interval is linear $(D_p^2(1\ \mathrm{g};\, V) \sim b_1 \,.\, b_2 \,.\, b_3 = V)$, in the second interval grows proportionally to $b_{31} \,.\, b_2 \,.\, b_3/b_3 = (b_{31}/b_3)^2 \,.\, b_1 \,.\, b_2 \,.\, b_3 = (V_1/V)^{2/3} \,.\, V = V \approx \,.\, V^{1/3}$ and thus proportinoally to $V^{1/3}$. In the third interval $D_p^2(1\ \mathrm{g};\, V) \approx V^{-1/3}$ and in the fourth interval $D_p^2(1\ \mathrm{g};\, V) \approx V^{-1}$.

Examples of the dependences of the variance $D_p^2(1\ \mathrm{g};\, b_1 \,.\, b_2 \,.\, b_3)$ on the volume $V = b_1 \,.\, b_2 \,.\, b_3$ computed for various ratios $b_1 : b_2 : b_3$ for selected periods $\alpha_1 = 8$ (length units), $\alpha_2 = 9$ and $\alpha_3 = 10$ are shown in Figs 2 and 3. Each curve starts from origin (point *0*) and is designated by a digit at its heighest point. For orientation the curve *6* in Fig. 2 passes through points *0, A, 6, B, C*.

From comparison of the curves plotted in Fig. 2 one can derive a rule for the determination of optimum orientation of particles in the compact material. For graphs *1*–*6* the shape of the particle is identical, the edges are in the ratio 8 : 9 : 10. The curve *1* corresponds to such an orientation of particles in the compact material when the ratio $b_1 : b_2 : b_3$ is the same as that of the periods of the fluctuation field $\alpha_1 : \alpha_2 : \alpha_3$ (*i.e.* 8 : 9 : 10). In this case, as may be seen, the second and the third interval dissappear as the condition $b_i \doteq \alpha_i/\pi$ becomes effective for $i = 1, 2, 3$ simultaneously (and hence $V_1 = V_2 = V_3$). This curve corresponds to the approximation used in the third paragraph (substitution of Eq. (*4*) by (*12*)) and as such is identical with the curve *p* in Fig. 1.

The graphs *2*–*5* correspond gradually to the orientations given by the ratios 8 : 9 : 10, 9 : 10 : 8, 10 : 8 : 9. The curve *6* corresponding to the orientation given by the ratio 10 : 9 : 8 has in the sections *0*–*A* and *B*–*C* the course identical with curves

1–5. In the section *A–B* it reaches minimum values in comparison with the others (in section *A–6* together with the curve *5*, in section *6–B* common with the curve *4*. The curve *6* thus corresponds to optimum orientation of particles of given shape. The corresponding ratio 10 : 9 : 8 thus means that optimum orientation of particles in the compact material is such whose shortest edge has identical direction with that of the longest period and the longest edge has the direction of the shortest period.

Thus conclusion confirmed also by the curves *7–9*, corresponding to particles shaped as slabs: The curve *7* corresponds to the orientation given by the ratios 1 : 2 : 2, curve *8* to the ratio 2 : 1 : 2 and curve *9* to 2 : 2 : 1.

The curves shown in Fig. 3 serve to find optimum shape of the particle. The curve *10* corresponds to particles shaped as a cube, curve *11* to a slab (2 : 2 : 1), curve *12* to parallelepiped (4 : 3 : 2), curve *13* to a rod (2 : 1 : 1), curve *14* to a thinner slab (3 : 3 : 1), curve *15* to parallelepiped (3 : 2 : 1) curve *16* to a slimmer rod (3 : 1 : 1).

Curves *10–16* point at the following conclusion: For particles of the same volume the variance $D_p^2(1\ g; b_1 . b_2 . b_3)$ is the smaller the farther away the shape of the particle from cube (*i.e.* the thinner the slab or the rod). In deciding between the slab and the rod for a definite material one has to take into consideration also the course of the remaining components of the variance of inhomogeneity, particularly that of $D_r^2(1\ g; V)$ as well as technical feasibility of preparing particles of the selected shape. Quantitatively, the rod-like shape may be regarded as suitable for smaller particles, the slab is more suitable for larger particles. For the case of the existence of a minimum on the course of the inhomogeneity variance $D_J^2(1\ g; b_1 . b_2 b_3)$ we shall have instead

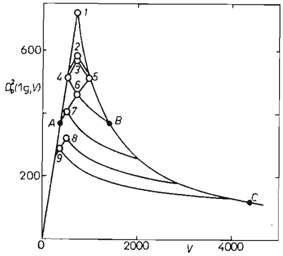

FIG. 2

Dependence of the Periodic Component of the Inhomogeneity Variance on Particle Volume for Various Orientations of the Particles in the Original Compact Material

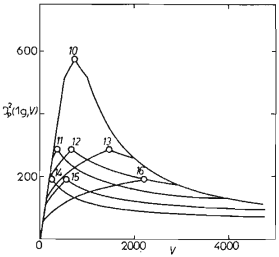

FIG. 3

Dependence of the Periodic Components of the Inhomogeneity Variance on Particle Volume for Various Shapes of the Particle of Eq. (*15*) (derived from comparison of Eqs (*10*) and (*13*)) a more general condition starting from Eqs (*10*) and (*19*)

$$D_r^2 < D_p^2(\omega_0)\, C(V_1/V_2)^{2/3} . \tag{24}$$

In words this means that at the terminal point of the second interval ($V = V_2$) the curve *p* has a larger value than curve *r*. For slabs, as may be apparent from Fig. 3, the ratio V_1/V_2 has a larger value than that valid for rods.

In the examination of the dependence on nonperiodic components of the inhomogeneity variance on the shape of particles and their orientation within the material we shall proceed similarly as in the previous case. The results in the search for the optimum shape of the particle and its optimum orientation, however, play a minor role and we shall therefore confine ourselves to a brief description only. For a given shape of the sample particle one can find from $D_n^2(b_1 . b_2 . b_3)$ and $D_n^2(1\ \text{g}; b_1 . b_2 . b_3)$ the magnitudes of the volumes V_1, V_2, V_3 (for V_n we have from curve *n* in Fig. 1 $V_1 \leqq V_n \leqq V_3$) delimiting the dependence of $D_n^2(1\ \text{g}; V)$ on V to four intervals: for $V \leqq V_1$ the variance $D_n^2(1\ \text{g}; V)$ grows with the volume V as in curve *n* (Fig. 1) *i.e.* proportionally to V, for $V_1 \leqq V \leqq V_2$ $D_n^2(1\ \text{g}; V)$ grows with $V^{2/3}$, while for $V_2 \leqq V \leqq V_3$ with $V^{1/3}$ and, finally, for $V \geqq V_3$ $D_n^2(1\ \text{g}; V)$ remains constant.

From a series of curves shown in this work for various particle shapes a general tendency has been found leading to the conclusion that without more detailed information about the inhomogeneity of the compact material the inhomogeneity of the granular material prepared from the former can be approximately minimized provided that the two following stipulations are met simultaneously: *i*) the volume of the particle is as small as possible and *ii*) their shape differs as much as possible from that of a cube (*i.e.* slabs or rods).

A substantial role may be played by the orientation of particles in the original compact material; the knowledge of the structure of the material, however, is indispensable. The general conclusion of this work (taking into consideration only periodic components) is as follows: Optimum orientation of particles in the original compact material is such when the shortest edge of the particle has the direction of the longest period of the periodic component of concentration fluctuation and the longest edge the direction of the shortest period.

Apart from these qualitative conclusions the paper enables also quantitative conclusions to be made on the basis of optimization of parameters of the granular material using formulas and approaches presented in this work. A prerequisite for their application, however, is the knowledge of the parameters of inhomogeneity of the original compact material in the sense of ref.[2] of which this paper is an extension.

REFERENCES

1. Stacheev Ja. I., Kuznecov J. I.: Zavod. Lab. *31*, 1 (1970).
2. Boháček P.: This Journal *42*, 2982 (1977).
3. Boháček P.: This Journal *42*, 3003 (1977).
4. Novák J., Bičovský K.: Hutn. Listy *26*, 66 (1971).
5. Boháček P., Jurek K.: This Journal *43*, 545 (1978).

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