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## **INTERPARTICLE DISTANCES IN THE CROSS SECTION OF GRANULES WITH DIFFERENT GRAIN SIZE DISTRIBUTIONS**

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Results of microscopic studies of interparticle distances in granules with different grain size distributions are presented in the paper. It was found on the basis of the analysis of luminance profiles of the image of the granule cross section that the granule consisted of multi-layer aggregates of grains placed at random in the granule volume. The range of changes in interparticle distances is related to the size of material grains.

*Key words: granulation, granule, structure, configuration, distribution of particles*

### **INTRODUCTION**

A raw material for the process of agglomerative granulation is a fine-grained solid whose particles under the influence of binding liquid and mechanical interactions form cohesive agglomerates – granules. A granule is a system of  $n$  particles which are combined by the forces of mutual interactions (Strelkov 1982). It has a finite potential energy of potential interactions between the  $i$ -th and  $k$ -th particle belonging to the system. The extent and type of interactions depend on the following factors: the method of granulation, chemical and physical properties of a solid body and binding liquid, as well as on the process conditions. During the process, particles of fine-

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grained solid with batch density corresponding to their position, transfer to the state resulting from the density of formed granules. The models of particle growth developed so far (Batterham et al. 1990) take into account neither changes of the density nor the structure of the granules being formed. In general, a predominating principle is to assume a defined mechanism of growth for a given method of granulation.

Many models of particle beds developed until present (Ouchiyama and Tanaka 1986, Yu and Standish 1987, Nolan and Kavanagh 1992) refer to systems consisting of a few components and built from spherical particles. The simplest elementary model of a granule is a system of two spherical particles of the same size, combined by interparticle forces. At the steady, average force of interactions between the two particles, the potential energy of the binary system depends on a distance from each other, so the determination of particle position within a granule is a vital problem.

Particles in a granule (the system of grains limited by the interfacial area) are arranged in some characteristic pattern. The closest neighbours of each grain reveals some order which decides about the granule microstructure. This order should refer both to the number of neighbours, as well as their location. In the case of grains of the same size the order should be the same for each grain. In a polydisperse system of particles, one may expect a significant differentiation depending on the particle size. The probability of finding a particle in the given point of granule volume depends on assuming a different position by another particle.

A probabilistic relationship between the position of particles can be described quantitatively by means of a bivariate distribution function of random variable  $X$ , which can assume only two values  $x_1$  and  $x_2$  with positive probability:

$$P(X = x_1) = p \quad (1)$$

$$P(X = x_2) = 1 - p \quad (2)$$

where  $0 < p < 1$ .

If a sphere of radius  $r$  limited by a layer of thickness  $dr$  is distinguished in a spherical granule, the number of particles in the layer surrounding the particle placed inside the granule is

$$dN = 4\pi(N/V)F(r)r^2 dr \quad (3)$$

where:  $N$  – number of particles in volume  $V$ , and  $F(r)$  – distribution function.

The aim of the study was to determine interparticle distances, the character and the parameters of distribution in granules formed from materials with different particle

size distribution. The values were determined on the basis of particle configuration in the system during experimental microscopic studies.

## EXPERIMENTAL

The structure was investigated in a set-up equipped with a reflecting microscope Olympus SZ11, with a camera, light source and a computer Pentium 133 equipped with an image analysis system "Lucia".

Granules formed from dolomite with three ranges of particle size (containing grains 0.16-20, 0.16-60 and 0.16-100  $\mu\text{m}$ ) and from glass balls (19-76  $\mu\text{m}$ ) of mean diameter 50  $\mu\text{m}$  were tested. The tested granules were ground on a flat screen to approximately half their diameter in order to form a flat intersection. The obtained granule was the subject of microscopic studies.

A fragment of the intersection field illuminated by a two-point light source was observed in the microscope and next, by means of a camera transmitted to the computer, where it appeared in the form of a rectangular image on the screen.

The position of five analysed images (in the form of rectangles 0.85 mm long and 0.65 mm high) along the longer axis of the intersection area is shown in Fig. 1.

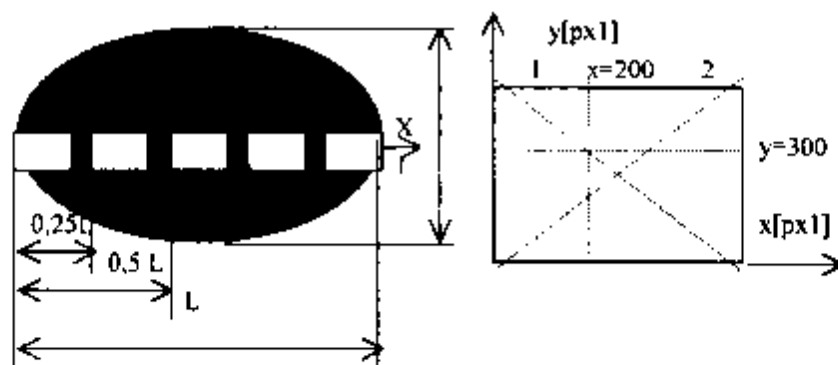


Fig. 1. Position of fields, coordinate axis and measuring points

Two of them include grains in the outer layer ( $X = 0$ ,  $X = L$ ), one in the middle and two others at a distance  $X = 0.25 L$ . In the measuring points of constant coordinates  $y = 300 \text{ px}$  and  $x = 200 \text{ px}$  the local image brightness (in the form of an abstract number) was determined. For the applied calibration the distance of measuring points was 0.00119 mm. One measuring length included a hundred points. The defined values of brightness of particular image points corresponded to the characteristic positions of grains and interparticle gaps.

## RESULTS

The dependence of brightness on position defines the cross section profile which is given in the form of a diagram or numerical data. Figure 2 presents an example of the profile of 0.16-60  $\mu\text{m}$  dolomite granule intersection area, referring to the image located at a distance of  $0.25L$  from the intersection edge.

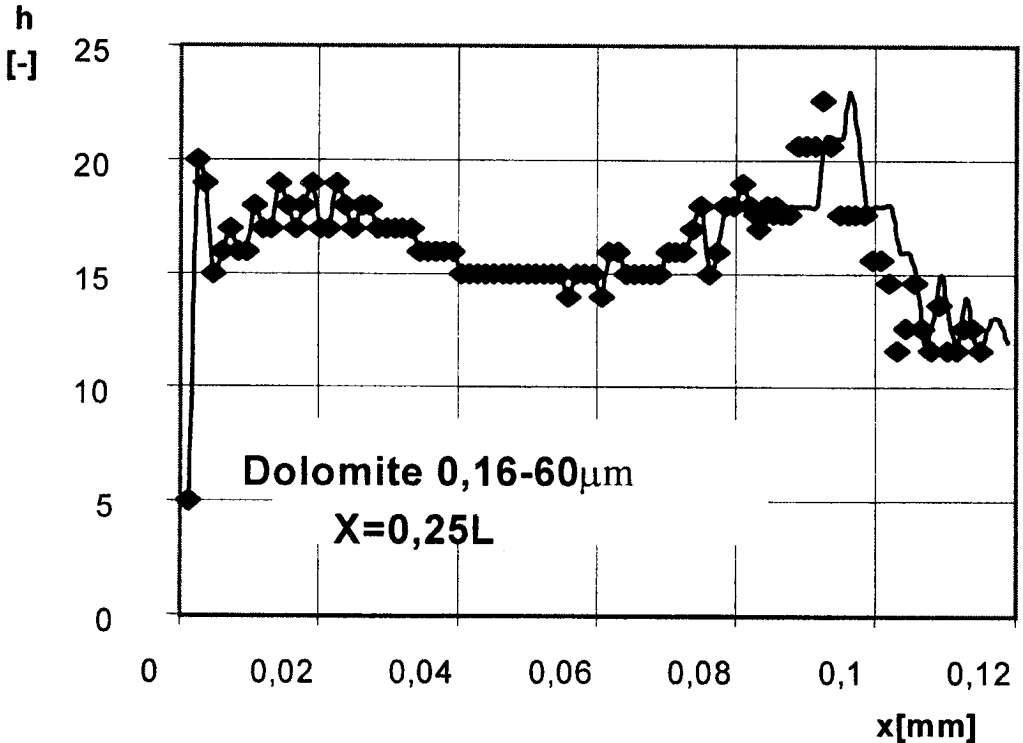


Fig. 2. Profile of 0.16-60 mm dolomite granule intersection area

The profile of glass ball granule intersection drawn for axis  $x$  is shown in Fig. 3. On the basis of these data the distances between peaks of a profile curve were determined. Each peak of the profile curve corresponds to the local brightness maximum and, in fact, provides an evidence of the presence of the next solid grain in this place. Diagrams of the dependence of interparticle distances on the position of solid particles along measuring axes  $x$ ,  $y$  were drawn. Examples of these dependences are illustrated in Fig. 4.

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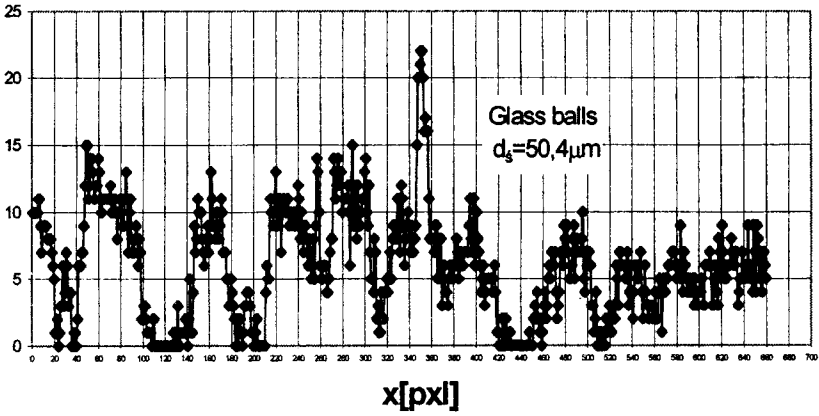


Fig. 3. Profile of granule intersection along the horizontal axis of the image

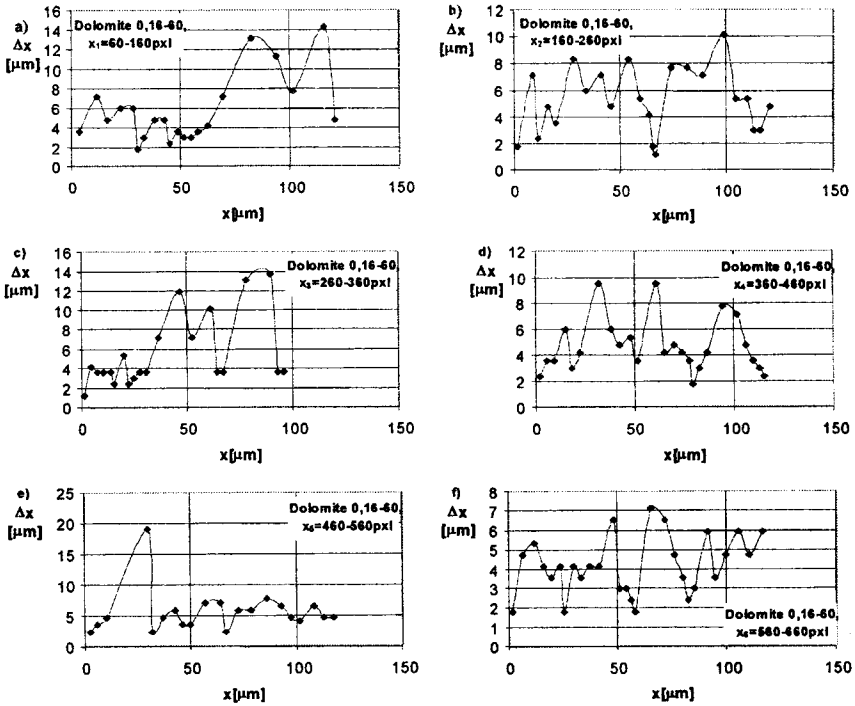


Fig. 4. Dependence of the interparticle distance on the position for particular measuring lengths

For a given measuring length the mean interparticle distance  $\Delta x_{\bar{x}}$  was calculated:

$$\Delta x_{\bar{x}} = \frac{\sum_{i=1}^n \Delta x_i}{n} \quad (3)$$

standard deviation  $s(\Delta x)$

$$s(\Delta x) = \sqrt{\frac{\sum_{i=1}^n (\Delta x_i - \Delta x_{\bar{x}})^2}{n-1}} \quad (4)$$

and density distribution  $q(\Delta x)$ .

$$q(\Delta x) = \frac{n_i}{\Delta(\Delta x) \sum_{i=1}^n n_i} \quad (5)$$

The dependence of mean interparticle distance in the tested granules on the position along the intersection axis is shown in Fig. 5.

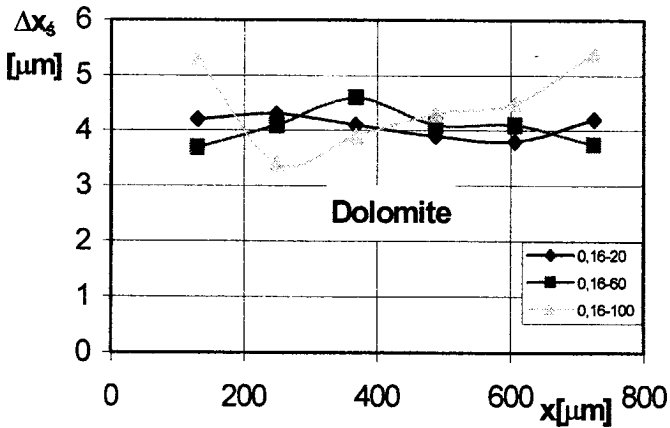


Fig. 5. Dependence of mean interparticle distance on the position of measuring length in the measuring field  $X = 0.5L$

The mean distance between solid particles  $\Delta_s$  was calculated for particular measuring fields as follows:

$$\Delta_s = \frac{\sum_{k=1}^6 \sum_{i=1}^n \Delta x_i}{\sum_{k=1}^6 \sum_{i=1}^n n_i} \tag{6}$$

The distribution of interparticle distances for various field in the granule intersection is illustrated in Fig. 6. The values of dimensionless distance of the measuring field from the intersection edge presented in the form of quotient X/L are represented on the axis of abscissae, and on the axis of ordinates there are mean values of interparticle distances.

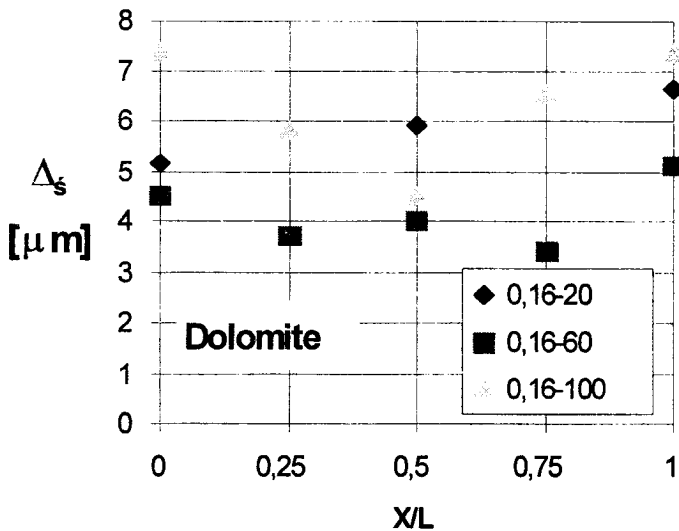


Fig. 6. Distribution of interparticle distances along the granule intersection axis

The functions of distance density distributions in the measuring fields  $X = 0.5L$  in the intersections of tested dolomite granules are shown in Fig. 7. Figure 8 shows the functions of density distributions for distances in the field  $X = 0.5L$  along its diagonals and axis  $x$  and  $y$  calculated for granules of glass balls.

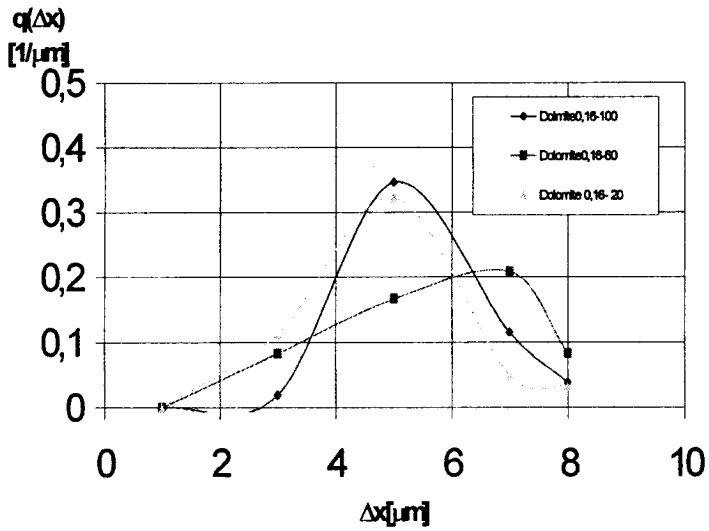


Fig. 7. Diagrams of density distributions of interparticle distances in dolomite granules

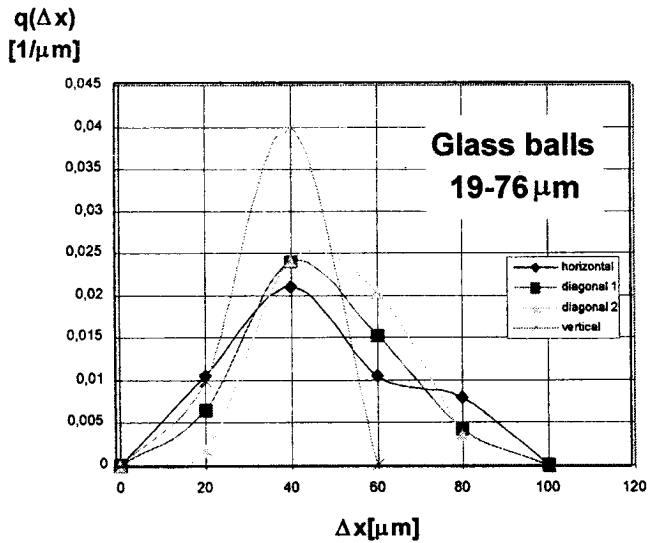


Fig. 8. Distance distributions on model granules made from glass balls



A comparison of density distributions of distances determined for particular measuring lengths of the field  $X = 0.5L$  for 0.16-60 dolomite granules is shown in Fig. 9.

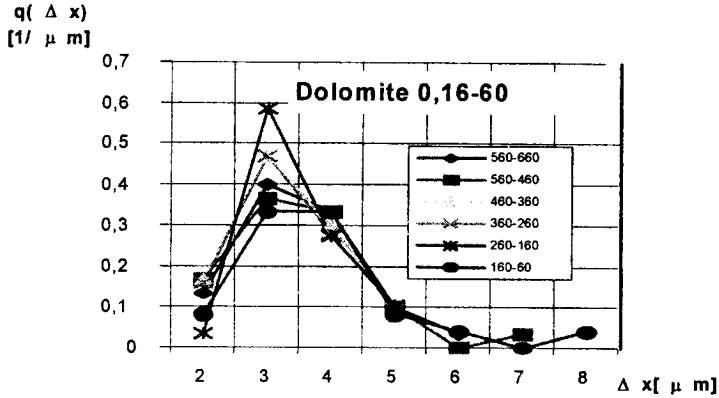


Fig. 9. Distributions of interparticle distances in measuring lengths of the field  $0.5L$  in the 0.16-60 dolomite granule intersection

### CONCLUSIONS

The following conclusions can be drawn on the basis of results obtained:

The profiles of surface brightness determined by the microscopic method suggest that granules have a discrete structure and can be used in the studies of microstructure.

A granule is formed as a result of coalescence of multilayer aggregates of particles (microgranules) which emerged at the stage of nucleation and after completing the process constitute local centres of particles concentration.

A multilayer (most frequently three or four peaks of the profile separate the brightest from the darkest point of the profile) structure of microgranules provides an evidence of a limited range of liquid penetration in the interparticle space.

The range of changes of interparticle distances is related to the range of grain size of the raw material. The largest distances occur in 0.16-100 dolomite, while the smallest in 0.16-20 dolomite.

The density functions of interparticle distance distributions for tested granules, similar to normal distributions, provide the evidence of a random character of their distribution in space.

The distance between particles changes periodically along the measuring field of the granule cross section and depends on the range of particle sizes of granulated material.

#### REFERENCES

- STRELKOV S.P. (1982), *Mechanics*, Mir Publishers, Moscow.
- BATTERHAM R.J., CRASS M., THURLBY J.A. (1990), *Review of Modeling in Agglomeration System*, ICHIME – 5th International Symposium on Agglomeration, Brighton, 569.
- OUCHIYAMA N., TANAKA T. (1986). *Porosity estimation from particle size distribution*, Ind. Eng. Chem. Fundamentals 25, 125.
- YU A.B., STANDISH N. (1987), *Porosity calculations of multi-component mixtures of spherical particles*, Powder Techn. 52, 233.
- NOLAN G.T., KAVANAGH P.E. (1992), *Computer simulation of random packing of spheres*, Powder Techn. 72, 149.

**Gluba T., Kochański B.**, Odległości międzyziarnowe w przekroju granulek substancji o różnym składzie ziarnowym, *Fizykochemiczne Problemy Mineralurgii*, 34 (2000), 57-66, (w jęz. angielskim).

Badania struktury prowadzono na stanowisku pomiarowym wyposażonym w mikroskop światła odbitego Olympus SZ11 za pomocą komputerowego w systemu analizy obrazu „Lucia”. Badano granulki utworzone z dolomitu o trzech zakresach wielkości cząstek (zawierającego ziarna 0,16-20, 0,16-60 i 0,16-100 $\mu\text{m}$ ) oraz ze szklanych kulek (19-76 $\mu\text{m}$ ) o średniej średnicy 50 $\mu\text{m}$ . Przeznaczone do badań granulki, ścierano na płaskiej siatce do mniej więcej połowy w celu utworzenia płaskiego przekroju średnicowego. Analizowano pięć obrazów (w kształcie prostokątów o długości 0,85mm i wysokości 0,65mm) położonych wzdłuż dłuższej osi pola przekroju. Dla stosowanej kalibracji odległość punktów pomiarowych wynosiła 0,00119mm. Jeden odcinek pomiarowy obejmował sto punktów. Określone wartości jasności poszczególnych punktów obrazu odpowiadały charakterystycznym miejscom. Na podstawie tych danych określano odległości między pikami krzywej profilu. Sporządzono wykresy zależności odległości pomiędzy cząstkami ciała stałego od ich położenia wzdłuż osi pomiarowych oraz funkcji gęstości rozkładów odległości w polach pomiarowych  $X=0,5L$  w przekrojach granulek badanych dolomitów i kulek szklanych. Stwierdzono, że granulka składa się z kilkuwarstwowych agregatów ziaren rozmieszczonych losowo w jej objętości. Zakres zmian odległości międzyziarnowych związana jest z zakresem wielkości ziaren surowca.

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