Simulation of spatial ordering of powder mixture particles when packed in square matrices using an example of monodisperse balls

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Abstract. The peculiarities of self-ordering of large particles of powder mixture components with layer-by-layer pressing of long billets used in material production by the method of self-propagating high-temperature synthesis (SHS) are studied using the example of structure formation at compaction of thin layers of a model system of monodisperse calibrated solid balls (1 mm in diameter). In unilateral axial compaction, the case of packing of thin layers of balls (containing 1–5 monolayers) in a square matrix (with a ratio of the transverse dimension of the matrix to the diameter of the ball ~14) is considered at low pressures that do not cause significant deformation of the balls. In the disperse system under study, a structural ordering of the particles occurs with a characteristic stepped view of the curve of the compact height dependence on the number of balls in the initial charge of balls. With an increase in the number of balls in the charge, the density of thin layers changes periodically during the compaction. In the compacts, fragments of structures in which the particles are arranged orderly like regular packing of ideal balls appear. The square shape of the matrix provides the formation of fragments of rhombohedral and cubic packing. The influence of nonuniform distribution of the initial charge of balls inside the matrix on the structuring of thin layers during compaction is considered.

1. Introduction
The process of ordered arrangement of particles in the mold during the axial compaction of thin layers of powder multicomponent mixtures, including SHS ones, is still poorly known [1–3]. Investigation of the peculiarities of structural ordering in powder systems of different fractional compositions and control of this process are necessary to increase homogeneity of long compacted items and improve characteristics of synthesized materials as well as to study combustion in the model heterogeneous mixtures with ordered arrangement of components and reaction cells [4, 5]. At the beginning of the investigation, it is reasonable to reveal the compaction regularities in model systems of particle-balls in matrices of different cross sections. The aim of the work was to study experimentally the structuring features in a model system of monodisperse solid spheres when compacted in a square matrix. The influence of the cross-section shape of the matrix on the frequency of appearance of ordered structure fragments was studied; these structures consisted of the balls similar to ideal balls in regular packages [6].
2. Experimental
In the experiments, the samples consisted of calibrated steel balls of 1.000±0.005 mm in diameter were subjected to single-sided axial compaction in a square steel mold (square side \( L = 14.2 \) mm, \( L/d \approx 14 \)). The influence of uniformity of the initial distribution of the balls in the matrix on the process of compaction and ordering of spheres in a thin layer was studied (it is important for comparison with the compaction results of powder SHS mixture samples). Due to the fixation of the mold in a sloping position to the vertical axis (angles of slope \( \alpha = 0^\circ, 45^\circ, 90^\circ \)), the initial non-uniformity in the balls distribution by the matrix cross-section was achieved and compaction was carried out. At the chosen compaction pressures (\( P \)), the balls did not collapse or significantly deform – the density of the sample increased only due to the movement of balls during repacking and their spatially ordered arrangement in the volume bounded by the punch planes and the matrix base and walls. The number of the balls in the sample (\( N \)) varied from 0 to 900 with a step of 5 particles for the range \( N = 0–570 \) and with a step of 10 for \( N = 570–900 \). The height (\( H \)) of the compacted sample was registered as a packing of a ball layer. The sample contained from 1 to 5 monolayers of balls in height. For \( N = 1–900 \) the condition of \( H << L \) was satisfied. Relative density of the "thin layer" (\( \rho_o \)) was defined as \( \rho_o = \rho/\rho_s \) (\( \rho \) is the density of the ball; \( \rho \) is the ratio of the balls mass to their full volume \((V_s)\) in the "thin layer" \((\rho = \rho_s V_b N/V, \) where, \( V_b \) is the volume of one ball\)) and was calculated by the formula: \( \rho_o = \pi d^3 N/(6HL^2) \). In the compacted "thin layers" various packages occurred; at certain values of \( N \) they could have a structure close to that of fragments of regular dense packages of ideal balls [6–9]. It was expected that the chosen shape of the matrix cross section (square) would stimulate the formation of fragments of the cubic package.

3. Results and discussion
Dependences of the height \( H = f (N) \) and density \( \rho_o = f (N) \) of the compacted "thin layer" on the number of calibrated balls in the sample were obtained (figures 1 and 2).

Most of the experimental points fall on the curves \( H = f (N) \) with an increasing form for all the angles of slope. The characteristic feature of the experimental curves \( H = f (N) \) is a discontinuous, periodic increase in the height of the thin layer with a constant interval; it occurs after each stage of monotonic growth of \( H \). The compacted samples containing the same number of balls can have discretely different heights. In such samples, at the same values of \( N \), various structures of balls are obtained – dense (with a minimum of voids) and non-dense (with vast voids and vacancies for balls), the interval of \( N \) values between the steps on the curves \( H = f (N) \) is commonly kept constant.

In the square matrix, at the formation of thin layers, non-dense structures are observed more often, and the corresponding range of values of \( N \) is wider than that in the cylindrical matrices [9]. In the thin layer in the square matrix, up to four monolayers with dense structure can be contained, more than in the cylindrical matrices. In inclined matrices the non-uniformity of filling sample distribution is more pronounced, non-dense structures are more often realized. For \( \alpha = 0^\circ \), voids are observed in the upper monolayers of the structure, near the punch; at \( \alpha = 45^\circ, 90^\circ \) they are located at the lateral face of the matrix on the top.

Different sections of the stepped experimental curves \( H = f (N) \) and horizontally located arrays of experimental points in figure 1 can correspond to the thin layers with fragments structurally similar to regular arrangement of ideal balls – cubic, tetrahedral (1), octahedral (2), rhombohedral (3) (the names are given in accordance with the type of the unit cell [6, 9]). The heights \( H_n \) of the regular packages consisting of \( n \) monolayers are shown in figure 1 with horizontal dashed lines (1–3). A certain correspondence of \( H \) in the horizontal arrays of experimental points to the values of \( H_n \) is observed.
Figure 1. Dependence of balls layer height on their number: (a) $\alpha = 0^\circ$; (b) $\alpha = 45^\circ$; (c) $\alpha = 90^\circ$.

Figure 2. Dependence of relative density of thin layer on number of balls: 1 – experimental "envelope" curve (at $\alpha = 0^\circ$); 2–5 – lines corresponding to densities of loose structures of $1$–$4$ monolayers; 6–9 – calculated densities of structures in square matrices ($L/d \approx 14$) at consecutive arrangements of balls into tetrahedral (6), octahedral (7), rhombohedral (8) or cubic (9) packages ($\alpha = 0^\circ$–$90^\circ$ (a–c)).

The frequency histograms in figure 3 have a pronounced discrete form. The values of $N_c$ reach local maxima at certain values of $H$, which coincide or are close to vertical lines (dashed and dash-dotted lines) that correspond to the heights $H_n$ for regular packages of balls of $n = 1$–$5$ monolayers (tetrahedral (1), octahedral (2), rhombohedral (3)). The location of $N_c$ maxima near these lines indicates the possibility of formation of thin layers containing fragments of regular structures. When $\alpha$ changes from $0^\circ$ to $90^\circ$, the characteristic shape of the frequency histogram, the number of $N_c$ maxima and their location on the histogram, are basically preserved (figure 3). In the square matrix, when the balls are arranged along the faces near the vertex of the inner right angle in the matrix space, there are more favorable conditions for formation of fragments of such regular packages as rhombohedral, octahedral, and cubic ones. Due to the presence of right angles along the perimeter of the layer, the motion of the balls along the face, during movements and repackages, is limited to a perpendicular face, which increases the stability of the rhombohedral and cubic fragments in the corners of the matrix. In the volume of the thin layer removed from the corners and faces of the matrix, there are no obstacles for the appearance of fragments of stable and denser tetrahedral package.
Figure 3. Distribution of number of sample fillings $N_c$ according to compact heights:
(a) $\alpha = 0^\circ$; (b) $\alpha = 45^\circ$; (c) $\alpha = 90^\circ$. Vertical lines indicate the heights for regular packages of 2–5 monolayers of balls.

The relative density of thin layers is shown in figure 2. At all the values of $\alpha$ the dependences of the relative density on the number of balls $\rho_\circ = \rho_\circ(N)$ have a nonmonotonic form with periodical density maxima and minima.

The density extrema are similar to those for theoretical dependences for ideal balls. In figure 2, broad broken lines 6–9 correspond to the curves of the dependence of the density of thin layers of balls with regular structures (rhombohedral, octahedral, tetrahedral) on the basis of the square shape – for $L/d \approx 14$, which can be obtained by theoretical consideration of sequential package "ball-by-ball" of monolayers of a thin layer (only with minimal distortion of the structure) in the matrix. The period of the change in the amount of sample filling with which density maxima $(\Delta N_{\text{max}})$ appear $N_{\text{max}} = 180–205$ balls.

Relative frequencies of appearance of denser structures in the volume of the matrix in fractions as a percentage of $W_i = (J_i/J)\cdot100\%$ depending on the number of balls $N$ in the sample filling are shown in figures 4 and 5. The data correspond to the "envelope" curve $I$ in figure 2a, the value of $J = 18$ corresponds to the number of experimental points for the intervals $\pm \Delta N/2$ within the selected values of $N$ with $\Delta N = 30$ in the range of $N = 0–570$ and $\Delta N = 60$ at $N = 570–900$. The period of appearance of $W_i$ maxima is: $\Delta N \approx 190 \text{ – for dense structure (curves } I–3) \text{ and } \Delta N \approx 150 \text{ – for loose structures (figure 5).}$

The peculiarities of structuring in thin layers of powders must be taken into account when producing long multilayer compacts from powder mixtures by sequential layer-by-layer compaction. The choice of a single sample filling with a value corresponding to one of the maxima of the thin layer density will contribute to the formation of a long compact containing significant fragments with ordered structure of coarse-grained component of the mixture. In this case, homogeneity of the multilayer compacts and constancy of the density along the length will increase. The obtained results can be used in developing recommendations for increasing density, homogeneity, and ordering of structure of compacts and products obtained by SHS or powder metallurgy when the initial mixtures contain spherical components, granular powder mixtures, clad or composite particles.
4. Conclusion

In the matrix of square cross-section there is a structural ordering of particles in thin layers in a system of calibrated monodisperse balls, simulating powder SHS-mixture. Structuring manifests itself through the stepped course of the curve of the compact height dependence on the number of balls in the charge and is reflected in the periodic change in the density of the samples. The fragments with an ordered arrangement of balls in thin layers are similar in structure to regular packing of ideal balls. The square shape of the matrix section contributes to the formation of fragments of cubic and rhombohedral packing near the corners and walls of the form. The heterogeneity of the initial distribution of ball charges in the matrix provides the appearance of loose structures more often than that in cylindrical matrices [9].

The considered features of the structural ordering of particles in thin layers should be taken into account in preparation of long compacts by sequential layer-by-layer pressing of SHS-mixtures into molds with a square or rectangular cross-section.

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