PERIODIC CHARACTER OF THE DENSITY–CHARGE MASS RELATIONSHIP IN PRESSING THIN LAYERS OF SOME POWDERS

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The emergence of ordered structures of powder particles in pressing thin layers of the Ti + 2B mixture is considered. The periodic dependence of the pressing density on the batch mass and the steplike dependence of the density on the number of compacting shocks is obtained.

The present study is devoted to investigating the structural self-ordering in pressing thin layers of some powders. The regularities of powder compaction using various methods of pressing predetermine such essential properties of end products as porosity, density uniformity, and strength [1-3]. Of particular interest is the process of pressing thin layers of a powder, since in these conditions its friction against the mold walls is actually absent, i.e., the final density is dominated mostly by properties of the powder per se. In a progressive pressing of fairly thin layers, it is practicable to produce quite dense long-measuring samples, which are at the same time highly homogeneous. The current study considered the regularities of compacting thin layers of the Ti + 2B powder, subsequently used to obtain titanium diboride by the SHS method [4]. For preparing a blend, we took titanium of grade PTK and amorphous boron of grade B99-A in a stoichiometric proportion (on a TiB_2 basis). In experiments, we employed two mixtures, viz., blend No. 1, which is a mixture of polydispersed titanium and polydispersed boron, and blend No. 2, which is a mixture of a thin fraction of the powder ($d_1 \simeq$ 63-100 μm) separated from titanium and polydispersed boron. The powders were subjected for 6 h to a dry mixing in a wobbling drum mixer. The powders were pressed in a die with the inner diameter D = 4.2 mm in a unit for impulse pressing of thin layers of the powder. The dependence of the pressing density on the mass of the powder charge in the die was studied. The powder batches (M) were weighted on the VLR-20G balance accurate to $1 \cdot 10^{-8}$ kg and varied from $5 \cdot 10^{-6}$ to $5 \cdot 10^{-5}$ kg. The pressing height (H) was varied from $5 \cdot 10^{-2}$ to 2 mm. The value of H was determined from readings of a micrometer (with a scale value of $2 \cdot 10^{-6}$ m) in immersing a punch into the die. The plunger mass ranged within 0.05-1 kg, and the height of the plunger fall varied over the range $2 \cdot 10^{-2}$-$6 \cdot 10^{-2}$ m. In all experiments, the punch mass equal to $1.6 \cdot 10^{-3}$ kg was much smaller than the plunger mass. From 1 to 1000 shocks (N) were delivered on the punch; in this case we first measured the pressing height after the punch was weighted with no shock (see Fig. 2, curve 1), and further on, after the first, fifteenth, etc. shocks with an interval chosen as the experimental purposes required. The density of the pressed sample was determined by the formula $\rho = 4M/\pi D^2 H$. The error in the determination of ρ constitutes about 2% for the batch of the mass $M = 5 \cdot 10^{-6}$ kg and decreases with increasing batch mass.

The dependence of the relative pressing density on the charge mass (Fig. 1, curve 1), obtained from the experiments on pressing of the powder of polydispersed titanium, is of traditional descending character. The curve for compacting the batches of the boron powder (Fig. 1, curve 2) is of similar form. Since the data given in Fig. 1 pertain to the experiments on pressing of the powders in one mold (D = const), plots of the pressing density as a function of the height of the original titanium or boron charge have the same form.

However, in pressing two-component blend No. 1 we discovered a qualitatively different, specifically, nonmonotonic character of the dependence of the pressed sample density on the charge mass. As is seen from Fig. 2, with increasing mass of the pressed powder, values of the pressure density pass through a series of maxima and minima with clearly reproducible parameters of their realization (a period and a magnitude). For curves 1-3 (Fig. 2), the mass period ΔM constitutes about $(10 \pm 1) \cdot 10^{-6}$ kg starting from the second maximum. Values of the linear period ΔH, calculated from the formula $\Delta H = 4\Delta M/\rho\pi D^2$, are equal, respectively, to 790, 460, and 370 μm. Such values of ΔH are two orders of magnitude larger than the dimension of boron particles entering into the composition of blend No. 1, and yet they are relatively close in magnitude to the diameters

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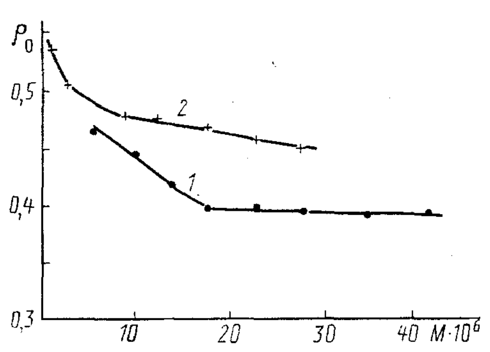

Fig. 1

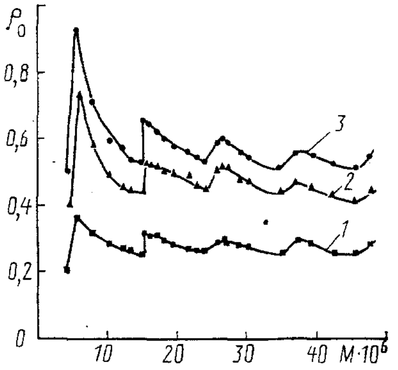

Fig. 2

Fig. 1. The relative pressing density vs the charge mass for N = 30, m = $6.84 \cdot 10^{-1}$ kg, and h = $4.3 \cdot 10^{-2}$ m: 1) titanium (PTK) powder; 2) boron powder. M, kg.

Fig. 2. The relative pressing density vs the mass of powder charge and the number of shocks delivered on the punch (Ti – PTK, B – amorphous B-99A); m = $6.84 \cdot 10^{-1}$ kg, h = $4.3 \cdot 10^{-2}$ m: 1) N = 0 (a nonshock action of the load on the punch); 2) N = 1; 3) N = 30.

of large titanium particles, whose fraction is noticeable in the polydispersed metal component of the powder mixture. According to data of the dispersion analysis, a fraction of 100-250 μm particles in the composition of the titanium (PTK) powder comprises about 70% of the total number of particles.

Pressing the mixture of boron and monodispersed titanium (blend No. 2) permits one to establish the relationship of the observed linear period ΔH to the characteristic dimension of the powder mixture, in the given case, to the diameter of titanium particles. Curve 1 in Fig. 3 shows the relation $\rho = \rho(M)$ for blend No. 2. A pronounced periodicity of the pressing density as a function of the charge mass is also observed in this curve. The period of variation in the height of the pressed layer, calculated from the above-given formula, is ΔH = 140-160 μm. To compare this value with the theoretical thickness of the blend "monolayer" L, wherein titanium particles form a single horizontal layer, we evaluated L from the equation

$$L \simeq \frac{d_1}{\rho_0^{1/3}} \left[1 + 2 \frac{A_2 \rho_1}{A_1 \rho_2} \right]^{1/3}. \tag{1}$$

For the case with $d_1 \simeq 100$ μm and ρ_0 = 0.5-0.6, the "monolayer" thickness is as large as L = 145-150 μm, which actually coincides with the experimental value of ΔH obtained above.

Thus, the foregoing enables one to assume that the revealed periodicity is linked with specific features of the behavior of titanium particles mixed with boron when these particles were stacked in layers during pressing. Since in the Ti + 2B blend the volumes occupied by the components are close in magnitude (V_1/V_2 = 1.12), and the pores take up about half the volume of the pressed sample, boron, by virtue of a small particle dimension ($d_2 \sim 1$ μm), possibly plays the part of a "dry" lubricant for relatively large titanium particles ($d_1 \sim 100$ μm), reducing the cohesion between them and thereby increasing their mobility relative to each other in repackings during pressing. In pressing very thin layers, whose heights are comparable or only by several factors exceed the particle dimension for a large fraction, this allows attainment of the most optimal (from the viewpoint of ensuring a high density of pressing) spatial structure of titanium particles for each specified mass of the blend charge. At the same time, pressing the titanium powder (without boron, i.e., without a "dry" lubricant), both polydispersed and monodispersed, under the same conditions, does not result in any appreciable deviations from the traditional monotonically descending character of the density-charge mass relation.

The emergence of the spatial structures of powder particles in pressing thin layers of the Ti + 2B blend may also account for the steplike run of the plot of the pressing density as a function of the number of pressing shocks (Fig. 4). This is especially evident on the curve section with N varying from 1 to 30 (Fig. 4a). "Shelves" on the curve apparently testify that the spatial structure of titanium particles, formed in the compacted powder, "holds a shock" for some time, till the variations accumulated in the pressing enable a new considerable displacement of titanium particles with the formation of a closer

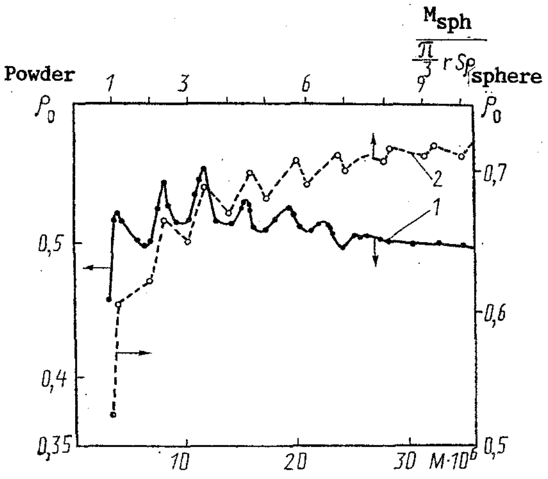

Fig. 3. Experimental dependence of the relative pressing density on the powder charge mass M for the Ti + 2B mixture, blend No. 2 (curve 1) ($m = 6.84 \cdot 10^{-1}$ kg, $h = 4.3 \cdot 10^{-2}$ m, $N = 30$) and the theoretical dependence of the relative density of the packing of spheres on their mass (2) (the curve maxima conform to a tetrahedral packing, and the minima to an octahedral packing).

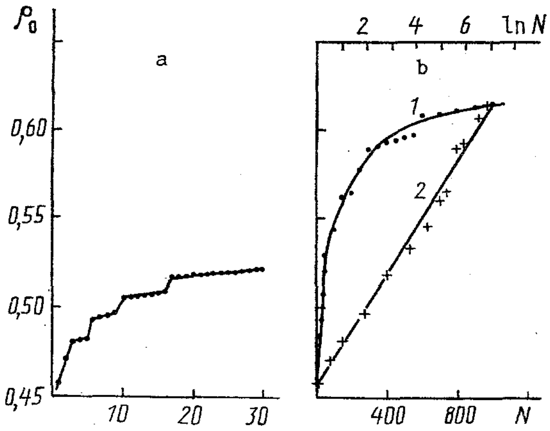

Fig. 4. The pressing density as a function of the number of compacting shocks: a) blend No. 1 [Ti(PTK) + 2B], $m = 6.84 \cdot 10^{-1}$ kg, $h = 4.3 \cdot 10^{-2}$ m, N = 1-30; b) 1, blend No. 1 [Ti(PTK) + 2B], $m = 6.84 \cdot 10^{-1}$ kg, $h = 4.3 \cdot 10^{-2}$ m, N = 1-1000; 2, the pressing density as a function of the number of shocks in a semilogarithmic scale.

packing of these particles. Obviously, with increasing pressing density the length of the "shelves" becomes ever more noticeable, whereas the magnitude of steplike density variations diminishes. Here, it is appropriate to note that the dependence of the pressing density on the number of shocks in the coordinates $\ln N - \rho_0$ has a near-linear form (see Fig. 4b) over a fairly wide range of parameters.

It is expedient to clarify the relation between the experimental results and the theoretical relations pertaining to the origination of various spatial structures in the packing of regular spherical particles of the same diameter. As is well known (see, for example, [5]), there exist four close packings of spheres (cubic, rhombohedral, octahedral, and tetrahedral), wherein (except in cubic) the average density increases asymptotically with increasing number of layers in the packing. In a cylindrical volume, i.e., in a space confined in two directions, for a prescribed discrete set of particles there may be a corresponding one of three ordered packings in K layers, viz., cubic, rhombohedral, or octahedral. Achieving a tetrahedral packing of the same number of layers K requires a greater number of spheres (by about $2/\sqrt{3}$ times) in each layer.

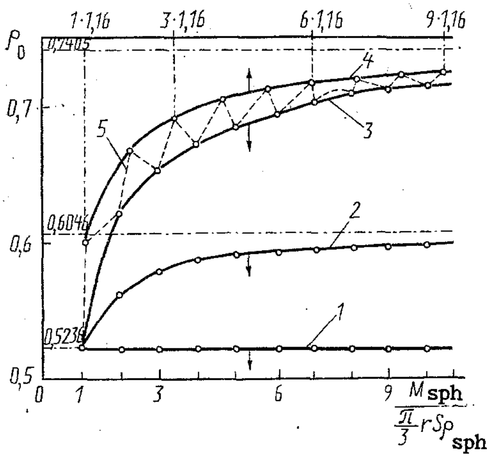

Fig. 5. The relative density of regular packings of spheres as a function of the reduced mass of spheres in a packing: 1, cubic packing; 2, rhombohedral packing; 3, octahedral packing; and 4, tetrahedral packing $[M_{sph}/(\pi/3)rS\rho_{sph}] = K$ for 1, 2 and 3; $M_{sph}/(\pi/3)rS\rho_{sph} = (2/\sqrt{3})K$ for 4 (K = 1, 2, 3,... is the number of layers in the packing); 5) schematic curve of the density variation with a "smooth" increase in the mass of spheres in a packing.

TABLE 1. Ratios of Pressing Densities at Maxima and Minima ρ_{max}/ρ_{min} for Various Numbers of Layers K

K	1	2	3	4	5	6
Experiment (mixture No. 2)	1,148	1,097	1,072	1,041	1,038	1,015
Theory (Fig. 5, curve 5)	1,152	1,071	1,060	1,045	1,043	1,035

Figure 5 shows the relative density for all four packings as a function of the number of layers in the packing in terms of the mass of the spheres M_{sph} (curves 1, 2, 3, and 4). For cubic, rhombohedral, and octahedral packings, $M_{sph} = (\pi/3)\rho_{sph}SrK$, and for a tetrahedral packing, $M_{sph} = 2/\sqrt{3}\ [(\pi/3)\rho_{sph}SrK)$. Thus, when the charge of spheres is compacted at appropriate discrete values of their mass, the packing densities can attain both a maximum, in the case of a tetrahedral packing, and a minimum (here we may anticipate, in principle, that one of the three packings, viz., octahedral, rhombohedral, or cubic, will form at the minimum). This can be exemplified by nonmonotonic "theoretical" curve 5 presented schematically in Fig. 5 and connecting the calculated maxima and minima of the density. The maxima correspond to the densities in a tetrahedral packing of K layers, and the minima to those in a tetrahedral packing of the same number of layers.

It is of interest to compare calculated curve 5 (Fig. 5) with experimental curve 1 (see Fig. 3). To present both curves in the same figure in the coordinates "density – number of layers" (theory) or "density – powder mass in a layer" (experiment), a scale of abscissas was chosen so that the period of the maxima of theoretical curve 2 (Fig. 3) corresponds to the period of the maxima $\overline{\Delta M}_{max} \simeq 4.06$ mg for experimental curve 1 (Fig. 3). It is evident from comparing the curves that the ratio of the averaged periods for the emergence of the density maxima $(\overline{\Delta M}_{max})$ to the period for the appearance of the minima $(\overline{\Delta M}_{min})$ is $\overline{\Delta M}_{max}/\overline{\Delta M}_{min} \simeq 1.1$ for the experimental curve and $\overline{\Delta M}_{max}/\overline{\Delta M}_{min} = 1.16$ for theoretical one. From Table 1 that ratios of the densities at the maxima to those at the minima (ρ_{max}/ρ_{min}) for the experimental and theoretical curves have close values, and with increasing K vary nearly in like manner.

Thus, the comparative analysis of the experimental relation for the density of the pressed sample and of the theoretical model for close packings of spheres indicates that, in the pressed mixture, titanium particles surrounded by fine boron tend to form the spatial structure, approaching a tetrahedral packing of spherical particles at the curve maxima, and an octahedral packing at the curve minima. Experimental curve 1 (Fig. 3) conforms to the run of theoretical curve 5 (see Fig. 5) to a charge mass equal to about 25 mg, which corresponds to seven periods of the density variation. It is difficult to further reveal the periodic character of the behavior of the pressing density because the difference in the densities at maxima and minima decreases with increasing number of layers (this also holds for the theoretical model). In this case, the measurement accuracy is insufficient for a correct interpretation of the experimental data. Besides, with increasing number of layers, the effect of particle repacking in itself is possibly reduced, since here the specific energy of the shock, responsible for compaction of a single layer of particles, decreases. The reduction of the average density of the pressed sample with increasing charge mass attests to such an assumption, because the theoretical curve for the sphere packing is, on the contrary, characterized by an increase in the average density with growing number of layers, with tetrahedral and octahedral packings simultaneously tending to one asymptotic limit with respect to density ($\rho_0 = 0.74$ for $K \to \infty$).

The character of the curves given in Fig. 5 shows that the indicated dissipative structures originate in thin layers of the powder charges with fairly low energies acting on the system. As is seen from curve 1, the periodic character of the relation $\rho = \rho(M)$ manifests itself under the effects corresponding to compaction of the layer with no shock, with the energy of order $7 \cdot 10^{-3}$ J.* Applying considerably stronger shock loads (curves 2 and 3, energies equal 0.29 and 8.7 J, respectively) increases only the absolute value of the density, without changing appreciably either the period, or the relative value of the oscillation amplitude of the density. As is well known, precisely such features are inherent in the dissipative structure [6].

CONCLUSIONS

The periodic dependence of the pressing density on the charge mass, established using a titanium–boron mixture as an example, finds a satisfactory explanation in the possibility of the formation of ordered structures, made up of relatively large titanium particles surrounded by finely dispersed boron. This conclusion is confirmed by the qualitative and quantitative agreement between the experimental data and the density characteristics of regular sphere packings (tetrahedral, octahedral, etc.). The adopted model also accounts for the steplike form of the dependence of the pressing density on the number of compacting impulses.

NOTATION

d_1, d_2, particle diameter of titanium and boron powders; D, die diameter; m, plunger mass; ΔM, mass period in variation of pressing density; ΔH, linear period in variation of pressing density; ρ, pressing density; ρ_0, relative pressing density; ρ_1, ρ_2, theoretical density of titanium and boron; A_1, A_2, atomic weight of titanium and boron; V_1, V_2, specific volume of titanium and boron in the Ti + 2B mixture; L, thickness of the "monolayer" of blend particles; K, number of layers in the ordered packing of spheres; ρ_{sph}, sphere density; r, sphere radius; S, area of the base on which spheres are packed; $\overline{\Delta M}_{max}$, $\overline{\Delta M}_{min}$, average periods of the emergence of density maxima and minima; ρ_{max}, ρ_{min}, maximum and minimum values of density; W, energy of nonshock pressing; g, acceleration due to gravity; h, height of the plunger fall.

LITERATURE CITED

1. B. S. Mitin (ed.), Powder Metallurgy and Spray Coatings [in Russian], Moscow (1987).
2. I. G. Shatalova, N. G. Gorbunov, and V. I. Likhtman, Physicochemical Basics of Vibrational Compaction of Powder Metals [in Russian], Moscow (1965).
3. A. G. Merzhanov (ed.), Combustion Processes in Chemical Engineering and Metallurgy [in Russian], Chernogolovka (1975).
4. A. G. Merzhanov, Regularities and Mechanism of Combustion for Pyrotechnical Mixtures of Titanium and Boron. Prept. [in Russian], Branch of the Institute for Chemical Physics, USSR Academy of Sciences, Chernogolovka (1978).
5. M. A. Gol'dshtik, Transfer Processes in a Granular Layer [in Russian], Novosibirsk (1984).
6. A. Yu. Loskutov and A. S. Mikhailov, Introduction to Synergetics [in Russian], Moscow (1990).

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*The energy W expended on pressing with no shock was estimated by the formula $W = mg\Delta H_0$, where m is the charge mass, and ΔH_0 is the punch displacement by the action of the load with no shock.