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Modelling of granule formation process of titan- Magnetite powdered materials by the method of rolling

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ABSTRACT

The construction of complex model of titan-magnetite granule formation of powdered materials in drum granulators taking into account of anisotropy of structure and laminating on surface is considered. It has been noted that granule formation proceeds in some stages depending on relaxation time of embryo formation. On the base of model a graphic interpretation of process of laminating of powder on surface is cited. Rheological model of compact of granules under action of external deformation stresses allowing to estimate the change of porosity and density in time of arrive is presented. The comparison of calculation and experimental results for evolution of distribution of granules on sizes has been presented.

Introduction

The granulation processes of powdered materials find a wide use in chemical, pharmaceutical, metallurgy and food industry. The necessity of granulation of powdered materials and requirement to their quality allowed developing the various devices and apparatuses: mixing device with high external stresses, rotating drum apparatuses, apparatuses with pseudo-liquid bed and other constructions [1]. The numerous experimental investigations of granulation process in mixing devices-granulators [1–6] and in drum apparatuses [6–10] showed that the end size of granules is determined by a great number of parameters among of which is important to note a size of forming nucleus-embryo, size of particles of powder and drops of binding substance, conditions of agglomeration, properties of powder and liquid and from method of granulation. In researches [3,4,10,11–14] the influence of sizes of particles of drops of binding substances on formation and further growth of granules and on morphology of structure is studied. It should be noted that prediction of corresponding quantity of liquid (size of drop) for preparation of desired size of granules is very difficult, owing to the fact that besides factors indicated above, sizes of forming granules depend on adhesion properties of powder, on physical properties of liquid (viscosity, surface tension). Laminating thickness and conditions of completeness of granule structure are determined by moisture-capacity or wet ability of surface. In this connection, in works [13,14] the influence of sizes of drops on growth rate and formation of embryos in granulators – mixers have been investigated.

It is introduced the permeation time of liquid into layer of powder, so called penetration time and determined as

$$\tau_p = 1{,}35\frac{\upsilon_0^{2/3}}{\varepsilon^2 R_p}\frac{\eta_s}{\sigma_L \cos\theta_0} \quad (1)$$

where υ_0 – volume of liquid drop, R_p – pores radius, ε – porosity, η_s – liquid viscosity, θ_0 – wetting angle, σ_L – surface tension of liquid.

The important problem in industrial processes of granulation of powdered materials are establishment of function of distribution of polydispersity of granules on sizes which allows to determine the change of their average size on length of apparatus in practical calculations [2,15,16]. The works [2,8,12,15] have been devoted to the experimental investigation of formation of granules of polydisperse composition and distribution of granules on sizes connected with it, measure of their sizes and porosity. The experimental curves of distribution of granules on sizes in mixers – granulators showed their two-humped character of curve of distribution, maximums of which are determined in the field of embryo formation and in the filed of structure formation of granule. In drum apparatuses the most efficient are the description of evolution of function of probability of granule sizes distribution with use of Fokker-Plank differential equation [15,18] on the base of experimental data characterizing continuous laminating and granule growth.

The granulation processes are accompanied by compact, deformation and granules wear [2,4,19,20] leading to change of their size, degree of polydispersity and physical

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properties – density, strength and porosity $[8,10,11]$. In particular, in work $[11]$ the change of granule density depending on time and characteristics of drum apparatus has been presented as

$$\frac{\partial\rho}{\partial t}=10^{-3,5}\left(D/L\right)^{-1.9}\omega^{0,06}k^{-1}$$

where $\omega-$angular speed of drum rotation, $D,L-$diameter and length of drum, k – some parameter. A great number of empirical formulas on calculation of physical properties of granules has been presented in works $[2,9-11]$. It should be noted that granules density is determined as $\rho=\rho_d\left(1-\varepsilon(t)\right)$, where ρ_d-density of material, $\varepsilon(t)-$ granule porosity depending on time and on rheology of laminating, compact and deformation about which will be stated below. Thus, as follows from this formula, the change of density has been connected with change of granule porosity in time.

As a whole, the granulation process, at first sight seeming simple, is the very complex phenomenon including investigation and description of such phenomena as embryo formation, structural formation of skeleton of granule itself, rheology of compact, deformation, etc.

The purpose of this work is the construction and analysis of complex model of formation of granule and its compact as a result of rolling taking into account of anisotropy of structure of forming granules.

Mechanism and model of titan-magnetite granule formation

The mechanism of granulation of titan-magnetite powdered materials (Fe - up to 13-15%; TiO_2 - up to 2.3-3.0%; V - up to 0.6%, Mn - 0.7%) by the method of rolling is determined by the following stages: a) mixing of powder with drops of binding substance and formation of nucleus of granule. Embryo formation in granulation process is determined by character of capillary interaction in layer of particles of powder with liquid, size of drops of binding substance, number of contacts in unit of volume of material (coordination number) and relaxation time τ_p. In practical cases the large particles (retour) being in composition of initial powder; b) growth and formation of granules as a result of their rolling on surface of powder can be served as source of embryo formation. In this stage the sizes of particles of powder (laminating thickness) and sizes of particles of drops of binding substance, rolling rate play an important role. The end size of granules is determined by degree of distribution of liquid due to capillary forces in pores and content of liquid in volume of granule (moisture-capacity); b) compact of granules under action of deforming external stresses and own weight. As a result of granule compact of liquid containing in pores is embossed to surface which increases a laminating rate of powder; c) stabilization and strengthening of structure of granule as a result of strengthening of internal bonds between separate particles in volume of granule and stabilization of end form.

Ultimately, the realization of form of granule is determined by geometry of two factors: geometry of dynamics of motion of granule in the process of its rolling and geometry of anisotropy of strength and other properties, exactly its resistance to rubbing and deformation. Dynamics of process is included in rotation motion of granule on surface of powder and apparatus, and the rotation is realized in all directions as a result of which geometry of dynamics of rolling has symmetries of rotating ball or sphere, i.e. consists of infinite quantity of axes of symmetry of infinite order.

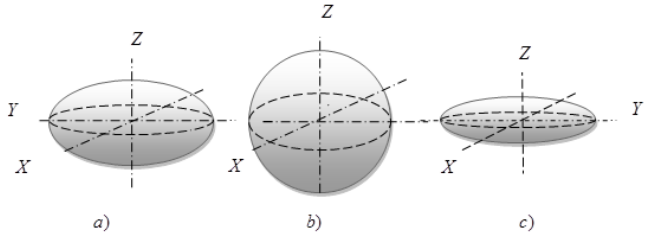

Fig1. Characteristic forms of granules (X,Y,Z – anisotropy axes)

Owing to in such geometry a dynamics of process of rolling in itself gives rounded form to granule (**fig.1**): oval (a), sphere (b), ellipse (c). On the other hand, the geometry of anisotropy of structure of granule is determined by rubbing, wear and deformation in all directions which creates the conditions for distortion and mixing of symmetry axes. However, in all cases of joint action of geometry of dynamics of rotation motion and anisotropy of structure, the form of granule approaches to rounded, exception strongly deformation ones. At the same time, as a result of action of various external and internal forces, granule loses a stability and is destroyed, losing in this case definite symmetry and form.

In spherical coordinates (r,θ,ψ) surface area of granule is determined as

$$S=\iint\sqrt{1+\frac{1}{r^2}\left(\frac{\partial r}{\partial\theta}\right)^2+\frac{1}{r^2\sin^2\theta}\left(\frac{\partial r}{\partial\psi}\right)^2}\,r^2\sin\theta\,d\theta\,d\psi \qquad (2)$$

where $\theta,\psi-$polar angles. If granule has a radius R, then as a result of laminating the radius of variable surface is increased per value of laminating thickness λ or $R+\lambda(\theta,\psi)$. If assume that $\lambda<<R$, then variable surface area taking into account (2) can be presented as

$$S\approx\iint\left[1+\frac{1}{2R^2}\left(\frac{\partial\lambda}{\partial\theta}\right)^2+\frac{1}{R^2\sin^2\theta}\left(\frac{\partial\lambda}{\partial\psi}\right)^2\right](R+\lambda)^2\sin\theta.d\theta d\psi \qquad (3)$$

In change of laminating thickness per value $\Delta\lambda$, surface area of granule will change per value

$$\Delta S=\iint\left[2(R+\lambda)\Delta\lambda+\frac{\partial\lambda}{\partial\theta}\frac{\partial\Delta\lambda}{\partial\theta}+\frac{1}{2R^2\sin^2\theta}\left(\frac{\partial\lambda}{\partial\psi}\right)\frac{\partial\Delta\lambda}{\partial\psi}\right]\sin\theta.d\theta d\psi \qquad (4)$$

The expression (4) can be considered as variation of surface area of granule on measure of change of laminating thickness. Integrating two last members on parts from 0 to π, we prepare

$$\int\frac{\partial\lambda}{\partial\theta}\frac{\partial\Delta\lambda}{\partial\theta}\sin\theta.d\theta=-\int\Delta\lambda\frac{\partial}{\partial\theta}\left(\sin\theta\frac{\partial\lambda}{\partial\theta}\right)d\theta$$

$$\int\frac{\partial\lambda}{\partial\psi}\frac{\partial\Delta\lambda}{\partial\theta}d\psi=-\int\Delta\lambda\frac{\partial^2\lambda}{\partial\psi^2}d\psi$$

Taking into account these expressions and assumed that on length Δl a change of surface area as a result of pure

laminating is equal to $\iint \lambda\Delta l d\theta d\psi$, the equation (4) will present as

$$\Delta S = \iint\left[\left(2(R+\lambda)-\frac{1}{\sin\theta}\frac{\partial}{\partial\theta}\left(\sin\theta\frac{\partial\lambda}{\partial\theta}\right)-\frac{1}{\sin^2\theta}\frac{\partial^2\lambda}{\partial\psi^2}\right)\Delta\lambda\sin\theta+\lambda\Delta l\right]d\theta\, d\psi \tag{5}$$

Change of granule volume is determined as

$$\Delta\upsilon = \int\int \Delta\lambda\left(R+\lambda\right)^2\sin\theta d\theta\, d\psi \tag{6}$$

Divided under integral expression (5) into (6), taking into account $\lambda << R$ we prepare under integral the expression corresponding to change of surface area of granule in relation to change of volume. Then passing to range we prepare the following expression

$$\frac{dS}{d\upsilon} = \frac{2}{R}-\frac{1}{R^2\sin\theta}\frac{\partial}{\partial\theta}\left(\sin\theta\frac{\partial\lambda}{\partial\theta}\right)-\frac{1}{R^2\sin^2\psi}\frac{\partial^2\lambda}{\partial\psi^2}+\lambda\frac{\partial l}{\partial\upsilon} \tag{7}$$

Multiplied and divided both parts into Δt and marked $d\upsilon/dt = \alpha$ and $V = \partial l/\partial t$, the equation (7) can be rewritten as

$$\frac{dS}{dt} = \alpha\left[\frac{2}{R}-\frac{1}{R^2\sin\theta}\frac{\partial}{\partial\theta}\left(\sin\theta\frac{\partial\lambda}{\partial\theta}\right)-\frac{1}{R^2\sin\theta}\frac{\partial^2\lambda}{\partial\psi^2}\right]+\lambda V \tag{8}$$

where V – mixing speed of granule determining geometry of dynamics. Thus, in equation (8) the first member reflects geometry of anisotropy of form, the second member – dynamics of motion of granule. In total case the equation (8) reflects asymmetric granule growth as a result of its rolling. For symmetric spherical granule assumed $S = \pi a^2$ (a – diameter of granule) we prepare more simple equation of change of average granule size as a result of its rolling

$$\frac{da}{dt} = \frac{2\alpha}{\pi a^2}+\frac{\lambda V}{2\pi a} \tag{9}$$

It should be noted that spherical form of granule corresponds to isotrope and uniform structure and is differed with invariability of strength and other properties of granule on various applications. The total solution of equation (9) is transcendental expression in connection of which we consider the private cases of its solution:

a) If $a << 4\alpha/\lambda V$, then equation (9) can be presented as

$$a^2\frac{da}{dt} = \frac{2\alpha}{\pi}, \qquad a(t)\big|_{t=0} = a_0$$

with solution

$$a(t) = \left(a_0^3+\frac{6\alpha}{\pi}t\right)^{1/3} \tag{10}$$

b) If $a > 4\alpha/\lambda V$, then equation (9) will be presented as

$$\frac{da}{dt} = \frac{\lambda V}{2\pi a} \tag{11}$$

with solution:

$$a(t) = \left(a_0^2+\frac{\lambda V}{\pi}t\right)^{1/2} \tag{12}$$

The expression (11) coincides with rolling equation in drum granulator cited in work [7], if $V = \omega R_B$ (where ω, R_B – angular speed and drum radius). Based upon solutions (10) and (12), the process of granule formation can be subdivided into two fields: $0 \le t \le \tau_P$, where structure formation of nucleus of granule is realized and $t > \tau_P$, where granule growth is realized due to laminating of powder on surface. Correspondingly, τ_P can be called relaxation time of embryo formation. Using equation (1) and assumed that volume of drop is equal to, $\upsilon_K = \pi d_K^3/6$ we prepare

$$\tau_P = 0{,}876\frac{d_K^2}{\varepsilon^2 R_P}\frac{\eta_S}{\sigma_S\cos\theta} \tag{13}$$

where R_P – pores radius. Actually, this equation corresponds to condition and filling time of capillary pores in ordered structures. It follows from equation (13) that with increase of average diameter of liquid drops, relaxation time of embryo formation is also increased proportionally square of diameter.

c) We consider stationary case of equation (8) at $t > \tau_P$, i.e.

$$\frac{\alpha}{R^2}\left[\frac{1}{\sin\theta}\frac{\partial}{\partial\theta}\left(\sin\theta\frac{\partial\lambda}{\partial\theta}\right)+\frac{1}{\sin^2\theta}\frac{\partial^2\lambda}{\partial\psi^2}\right] = \lambda V \tag{14}$$

If assume that laminating layer thickness is not changed relatively polar angle ψ, then the equation (14) can be rewritten as

$$\frac{\partial}{\partial\theta}\left(\sin\theta\frac{\partial\lambda}{\partial\theta}\right) = \frac{\lambda V R^2}{\alpha}\sin\theta$$

In view of insignificance of the second derivative $\partial^2\lambda/\partial\theta^2$, this equation can be written as

$$\cos\theta\frac{\partial\lambda}{\partial\theta} = \frac{\lambda V R^2}{\alpha}\sin\theta$$

Divided both parts into θ and integrating both parts, we have

$$Ci\theta = \beta.Si\theta \tag{15}$$

where $Ci\theta, Si\theta$ – integral cosine and integral sinus, $\beta = \dfrac{R^2V}{\alpha^{\partial\ln\lambda/\partial\theta}}$, θ – angle in radians. The expression (15) is equation of stationary laminating in the granulation process of powdered materials. In **fig. 2** the numerous trajectories of laminating of powder on surface of granules in various values β are presented. The presented curves give the graphical interpretation of laminating on surface of spherical granule.

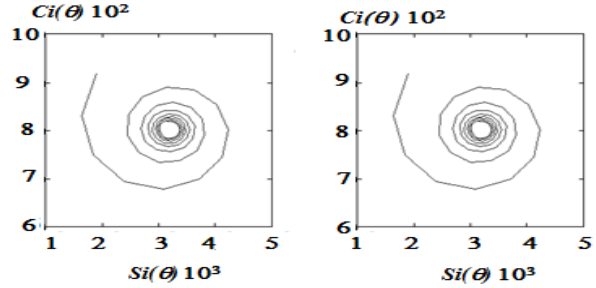

Fig.2 Geometrical interpretation of laminating granules in various β: a) $\beta = 0{,}2$; b) $\beta = 1{,}0$

Rheology of granule compact

Granule forming as a result of rolling under action of external deforming forces stipulated by interaction of granules between themselves and wall of apparatus is subjected to the compact owing to rearrangement of particles of powder in granule. Each particle of powder in granule contacts with multiply of others and in immediate closeness from contact between spherical particles the rounded capillary crack is formed, where binding substance is stayed. If assumed that sizes of particles of powder are approximately same then as a result of their agglomeration to single system the ordered structure with various coordination number N_k can be formed. In availability of a great number of fine-dispersed fractions as a result of compact more dense and durable structure of granule is prepared. Granule porosity in friable packaging $(N_k = 6)$ – $\varepsilon = 0{,}476$, but in sufficiently dense structure $(N_k = 12)$ – $\varepsilon = 0{,}259$ [20].

As a result of granule compact the hard phase (particles of powder) pours into pore compressing liquid in pores and excluding it on continuous reserved systems of pore canals. If the isolated reserved pores have been included in low penetrable or impenetrable medium of granule volume then a volume flow leads to increase of liquid pressure ΔP_f, thereby distorting form, engenders an anisotropy of internal structure. The character of external stresses acting to granule or to particles in granule is determined by availability of centrifugal forces appearing in drum rotation and mass forces stipulated by weight of above-lying layers and also a number of forces arising in collision of granules between themselves and wall of apparatus. On analogy with theory of elasticity [21], viscous deformation tensor rates are analogous to deformation tensor and the efficient coefficients of volume and shift viscosity are analogous to modules of thorough compression and shift. Thus, rheology of deformation and compact of porous granules are characterized by the efficient macroreological characteristics: shift viscosity coefficient ξ_S relaxation volume viscosity coefficient η_S, connected with volume flow of porous medium. Let us assume that in each local element of porous granule the macroscopic homogeneous stressed state is realized. According to the theory of elasticity the elastic volume deformations of skeleton of porous granule are connected with external deforming stress ratio

$$\frac{\Delta V_S}{V_S} = \frac{\sigma_D}{\eta_S}\Delta t \qquad (19)$$

where $V_S = 1/\rho_P$ – specific volume of porous medium, ΔV_S – change of volume of local element of medium, $\rho_P = \rho_d(1-\varepsilon)$ – poured density of granule, ρ_d – density of material of dense phase, $\sigma_D = \Gamma_D - \Delta P_f + g(\rho_d - \rho)z$ – deforming stress laying down from external stress, pressure drop inside of granule and weight of above-lying layers by thickness z. Differentiated (19) on t, following can be written

$$\frac{1}{1-\varepsilon}\frac{d\varepsilon}{dt} = -\frac{\sigma_D}{\eta_S} \qquad (20)$$

Passing from substantial derivative to local in equation (20), following will be obtained

$$\frac{\partial \varepsilon}{\partial t} + div\left(\varepsilon \overline{U}_S\right) = -(1-\varepsilon)\eta_S^{-1}\sigma_D \qquad (21)$$

where $\overline{U}_S$ – vector of particles motion rate in volume of granule as a result of its compact. In spherical coordinates this equation can be written as

$$\frac{\partial \varepsilon}{\partial t} + U_{Sr}\frac{\partial \varepsilon}{\partial r} + \frac{U_{S\theta}}{r}\frac{\partial \varepsilon}{\partial \theta} + \frac{U_{S\psi}}{r\sin\theta}\frac{\partial \varepsilon}{\partial \psi} = -(1-\varepsilon)\eta_S^{-1}\sigma_D \qquad (22)$$

The third and fourth members in the left part of this equation determine the uniform distribution of porosity on volume of granule engendering anisotropy of strength properties. For single-measure radical compact of granule the equation (21) is simplified to view

$$\frac{\partial \varepsilon}{\partial t} + U_{Sr}\frac{\partial \varepsilon}{\partial r} = -(1-\varepsilon)\eta_S^{-1}\sigma_D \qquad (23)$$

Expressed volume viscosity η_S through shift viscosity ξ_S as [22]

$$\eta_S = \frac{4}{3}\xi_S\frac{1-\varepsilon}{\varepsilon}$$

we prepare

$$\frac{\partial \varepsilon}{\partial t} + U_{Sr}\frac{\partial \varepsilon}{\partial r} = -\frac{3}{4}\varepsilon\xi_S^{-1}\sigma_D \qquad (24)$$

$$\varepsilon(t)\big|_{t=0} = \varepsilon_0$$

The solution of equation (24) can be presented as

$$\varepsilon(t) = \varepsilon_0 \exp\left[-\frac{3}{4}\xi_S^{-1}\sigma_D\left(t + \frac{r}{U_{Sr}}\right)\right] \qquad (25)$$

For average porosity on volume of granule can be written [19]

$$\varepsilon(t) = \varepsilon_0 \exp\left(-\frac{3}{4}\xi_S^{-1}\sigma_D\bar{t}\right) \qquad (26)$$

The equation (26) allows estimating the value of granule porosity in various moments of stay time taking into account compact and considering their values in calculation of physical characteristics. However if the initial and current values of granule porosity are known then from equation (26) the value of shift viscosity can be estimated as solution of reverse problem

$$\xi_S = \frac{3}{4}\frac{\sigma_D \bar{t}}{\ln \varepsilon_0 / \varepsilon}$$

For granulation process we estimate the value of shift viscosity in the following values: for friable structure of granule $\varepsilon_0 = 0{,}476$, for dense structure $-\varepsilon = 0{,}259$, granulation time $t = 480\,\text{sec}$ and deforming stress $\sigma_D \approx 100\frac{H}{м^2}$. In these data the value of shift viscosity is equal to $\xi_S = 6.10^4\,\frac{H.\text{sec}}{м^2}\cdot$

Assumed that change rate of granule surface as a result of compact is equal to $S^{d\varepsilon/dt}$, then laminating equation can be written as

$$\frac{da}{dt} = \frac{2\alpha}{\pi a^2} + \frac{\lambda V}{2\pi a} + \frac{a}{2}\frac{d\varepsilon}{dt} \qquad (27)$$

The expression (27) describes the process of granule formation of powdered materials in drum granulators. This equation includes three stages of granule formation: embryo formation $(t < \tau_P)$, formation of granules by laminating $(t > \tau_P)$ and their compact determining by the last member.

Taking into account the expressions (26) and (27), the equation of change of granule sizes will be presented as

$$\frac{da}{dt} = \frac{\lambda V}{2\pi\alpha} - \frac{3}{8}\frac{\sigma_D \varepsilon_0}{\xi_S}\exp\left(-\frac{3}{4}\frac{\sigma_D}{\xi_S}t\right) \qquad (28)$$

In view of insignificance $\frac{\sigma_D}{\xi_S} << 1$, linearing this equation we determine the approached solution (28) as

$$a \approx \sqrt{\beta_0 - \left(\beta_0 - a_0^2\right)\exp\left(-\beta_1 l + \beta_2 l^2\right)} \qquad (29)$$

where

$$\beta_0 = \frac{4}{3\pi}\frac{\lambda V \xi_S}{\sigma_D \varepsilon_0}, \qquad \beta_1 = \frac{3}{4}\frac{\sigma_D \varepsilon_0}{\xi_S V},$$

$$\beta_1 = \frac{9}{32}\left(\frac{\sigma_D}{\xi_S V}\right)^2 \varepsilon_0, \qquad a_0 - \text{average size of embryo, } l -$$

length of apparatus. With use of industrial data the estimation of coefficients will be β_0 = 1332.5; β_1 = $2{\cdot}10^{-3}$ and $\beta_1 = 3.3{\cdot}10^{-4}$. In **fig.3** the numerous solutions of equations (29) and experimental values of average granule sizes on length of drum apparatus have been presented. As follows from **fig.3**, in carrying out of conditions l > 60*cm*, granules size is rather decreased as a result of its compact and strengthening of structure.

. Geometrical interpretation of laminating granules in various β: a) $\beta = 0{,}2$; b) $\beta = 1{,}0$.

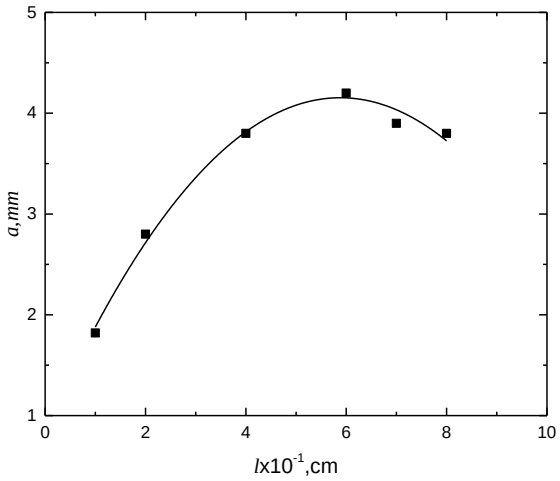

Fig 3. Variation of values of average granule size with length of drum (dots -experimental results, curve – mathematical result).

Strengthening, stabilization and fixation of structure of granule is carried out by two ways: a) use of surface coatings (capsulation) [23, 24]; b) strengthening of internal bonds between particles of powder in granule, by means of use of various additions to powder. Consequently, the main factor of stabilization of structure of granule is its strength and resistance to destruction and deformation as a result of action of external loads. Principally, the strength value can be estimated by the following polyempirical equation

$$\frac{d\ln\Delta}{d\tau_D} = f(k,\Delta) \qquad (30)$$

where Δ – strength or parameter characterizing strength of granule, τ_D – deformation time or parameter from which a strength depends, $f(k,\Delta)$ – function choosing on the base of experimental investigations, although formula rather analogous to equation (30) is presented in [24].

In conclusion we note that equation (8) and (27) can be also used for description of process of formation "snow ball" in availability of initial nucleus of laminating.

Experimental study of granulation of titan-magnetite concentrate produced with flux soda additive

Granulation of the resulting titan-magnetite powders was investigated in a laboratory tumbling granulator drum diameter 150 mm and length 750 mm present in **fig.4.**

The drum is a tin cylinder – 1, 150 mm in diameter and 750 mm in length with rods and pins - 2, non-slip granulated material on the inner surface of the drum. The back side of the drum head is attached to the flange - 3 fixed to the axis - 4 axis installed in two bearings, mounted respectively on two pillars - 5. axle gear planted - 6, by which the rotation of the gear - motor and 7 - 8 is transmitted axis of the drum.

Granule 6 installed on tiles and it is carefully lowered axis 2 with cup 1, fastened to its upper end. Axis free to move vertically in sleeve holder 3, fortified on a tripod 4. 1 cup slowly fall asleep until fraction crushing granules. The total weight of the axis of a cup and a fraction of the ultimate strength of the pellets kg cm^{-2}. We investigated the effect of time on the strength of natural drying quality of the resulting granules, so-called open time formed granules.

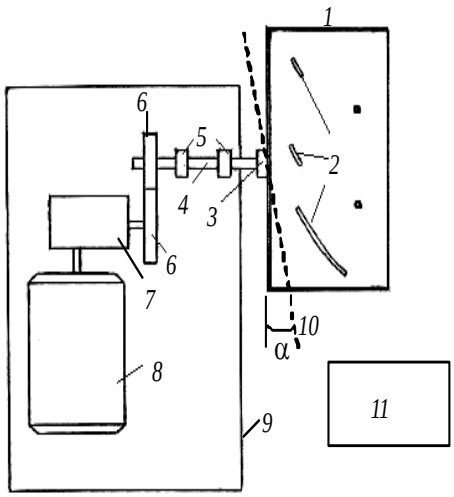

Fig.4. Drum granulator: 1 – drum, 2 - rods and pins, 3 - flange 4 - axis and 5 - bearings in supports, 6, 61 - gear and 7 - gear 8 - engine, 9 – stand, 10 - the angle α of inclination drum to the vertical plates 11 - hopper.

Determination of pellet strength according to the drying time after 1, 2, 3 or more hours are given in Table 1 below. Studies have shown that pellet strength increases with time, in particular after 3 hours, the most robust pellets appeared with a white coating on the surface of soda. Granule diameter 4mm 3 hours later collapsed at a load of 0.35 kg cm^{-2}, granule strength during natural drying increased almost 6 - 7 times as compared with fresh - 0.5 - 1 hour to the strength of 50-80 g cm^{-2}

Table 1. Increase in pellet strength versus time natural drying.

Granule diameter, mm	Granules strength, kg cm^{-2} natural drying, hour					
	0,5	1	2	3	24	48
3,5				0,200	0,650	0,65
4,0	0,050	0,08	0,130	0,250	0,75	0,85
4,5				0,300	0,80	0,90
5,0				0,350	1,0	1,25

After determining the strength of granule quality specified amount of water required for granulation. The amount of water measured on the one hand actually expended during spraying, and other refined water being in crude granule maximally saturated with water. This is the limit of saturation, the excess of which leads to blocking of granules, the formation of a solid mass of granulated material. For the first variant was weighed before the start of spray granulation of 45g sample after the end of the process, including the time of the additional rounding. The amount of water that went to the granulation and its natural loss amounted to 19g in%, based on the sample and the water consumed (just 64g), was 29.7%. The amount of water that went to the granulation (29.7; 29.3; 29.6; 29.4) averaged 29.5%, which is almost 3 times as big as the amount of water required for granulation of magnetite concentrate without soda. This is natural and is associated with the formation of crystalline soda binding considerable amounts of water. As noted above, the strength of the pellets at natural drying on the table during the first day significantly increases and then increases slightly. In connection with this we determined moisture content of pellets in natural drying desktop through 1; 2; 8 days. At the same time to clarify the amount of water that went to the measured moisture content of granules immediately after granulation. The data obtained are shown in Table. 2.

Table 2. Water loss at natural drying on a table with time.

Time	Humidity, %	Loss of water from initial quantity, %	Loss of water from residual, %
0	29,03	0	0
1	24,92; 85,9	4,11; 14,16	4,11; 14,16
2	17,95; 61,8	11,08; 38,70	6,95; 23,84
8	15,06; 51,9	13,97; 48,12	2,89; 9,96

As can be seen from Table 2 granule moisture is reduced by the eighth day almost 50% (48.12) of the amount of introduced water, but the strength of the pellets as discussed above is not reduced, but increases slightly. Thus, the in feed granulation 29% of water is quite optimal and its loss of 14% in a day or 38%, after 2 days does not reduce the strength of the granules.

Time evolution of granule size distribution

The base of stochastic description of granulation process of powdered materials is stochastic Focker-Plank differential equation recording as

$$\frac{\partial P(r,t)}{\partial t} = -\frac{\partial}{\partial r}\left[f(r)P(r,t)\right] + B\frac{\partial^2 P(r,t)}{\partial r^2} \qquad (37)$$

where $P(r,t)$ –density of function of granule distribution on sizes and at time, $f(r) = dr/dt$ – granule formation rate, B – stochastic diffusion coefficient, $r = a/a_0$ – stretch granule diameter. Based upon equation (28) and assumed $\sigma_D/\ \xi_S << 1$, granule formation rate in stretch form is determined as

$$\frac{dr}{dt} = \frac{m_R}{r} - kr \qquad (38)$$

where

$$m_R = \frac{V\lambda}{2\pi\alpha_0^2}, \quad k = \frac{3}{8}\frac{\sigma_D}{\xi_S}\varepsilon_0 .$$ Taking into account of expression (38) Focker-Plank equation will be written as

$$\frac{\partial P(r,t)}{\partial t} = -k\frac{\partial}{\partial r}\left[\left(\frac{m}{r} - r\right)P(r,t)\right] + \frac{B}{\alpha_0^2}\frac{\partial^2 P(r,t)}{\partial r^2} \qquad (39)$$

$t=0,\ P(r,t)=P_0$

where $m = m_R/k$. The solution of equation (39) by the method of division of variable ones will be presented as [15,16]

$$P(r,t) = r^{\theta}\exp\left(\frac{k\alpha_0^2 r^2}{2B}\right)\sum_{n=0}^{\infty} C_n L_n^{(\alpha)}\left(\frac{k\alpha_0^2 r^2}{2B}\right)\exp(-2knt) \qquad (40)$$

where

$$\theta = m_R\alpha_0^2/B, \quad \alpha = \frac{m_R\alpha_0^2 - B}{2B}, \quad L_n^{(\alpha)}$$ – Laquerre function.

$$C_n = \frac{\theta^{\frac{\theta+1}{2}}\int_0^{\infty} P_0(r)L_n^{(\alpha)}\left(\frac{k\alpha_0^2 r^2}{2B}\right)dr}{2^{\frac{\theta-1}{2}}\Gamma\left(n+\frac{\theta+1}{2}\right)m^{\frac{\theta+1}{\theta}}n!} \qquad (40a)$$

Table 3. Experimental distribution size of granule the apparatus .

Particle size, mm	0.1-0.2	0.2-0.4	0.6-0.8	0.8-1.0	1.0-1.2	1.2-1.5	1.5-2.0	2.0-2.5	2.5-3.0	3.0-4.0	4.0-5.0	5.0-6.0
L=10cm m_g, gr. ϕ	14.63 0.073	18.53 0.092	27.3 0.1365	29.75 0.198	27.80 0.139	22.40 0.110	18.00 0.090	11.20 0.056	5.850 0.030	1.950 0.0097	0.480 0.0024	0.0 0.0
L=20cm m_g, gr. ϕ	9.870 0.049	11.80 0.060	17.70 0.089	19.73 0.098	22.37 0.110	26.30 0.130	25.00 0.125	21.60 0.108	16.40 0.080	9.20 0.046	6.0 0.03	0.350 0.003
L=40cm m_g, gr. ϕ	4.680 0.023	7.80 0.039	13.30 0.066	14.80 0.074	17.20 0.0859	20.30 0.10	21.80 0.109	25.00 0.125	23.40 0,117	20.30 0.101	14.84 0.074	6.20 0.03
L=60cm m_g, gr. ϕ	6.600 0.0329	8.800 0.044	13.90 0.07	15.40 0.078	17.60 0.088	20.50 0.102	21.90 0.109	24.90 0.184	26.4 0.130	17.59 0.088	10.25 0.050	4.30 0.02
L=80cm m_g, gr. ϕ	2.890 0.014	4.340 0.0217	8.700 0.043	10.10 0.050	14.50 0.072	18.0 0.09	19.50 0.0975	20.60 0.103	28.80 0.144	30.78 0.154	22.40 0.112	12.450 0.0625

Where m_g is weight of granules of definite size in each sieve, φ is weight fraction.

The solutions (40) and (40a) characterize the evolution of function of density distribution of probability of granules on sizes and at time.

Conclusions

The analysis of granulation process of powdered materials stated in this investigation allowed describing the basic stages of granule formation: embryo formation, laminating as a result of drum rotation and granule growth, compact and creation of durable structure of granules (28) and (30). The models (8) and (22) consider the motion dynamics and anisotropy of form and strength properties. With use of stochastic Focker-Plank equation the evolution of function of granule distribution on length of apparatus and on sizes as the equations (40) and (40a) has been determined. The asymptotic value of distribution at $t \to \infty$ is prepared from solution (40), taking into account Laquerre function as

$$P(r) = C_R r^{\theta} \exp\left(-\frac{k\alpha_0^2 r^2}{2B}\right) = C_{PR} a^{\theta} \exp\left(-ba^2\right) \quad (41)$$

$$C_{PR} = 2a_0^{-\theta}\left(\frac{\theta}{2m}\right)^{\frac{\theta+1}{2}}, \quad b = k/2B$$

Thus, as follows from equation (41) in large stay time of granules in apparatus the range distribution which doesn't depend on initial distribution is established. In large values of granule compact rate such state is reached in fewer values of *t*. In practical calculations the use of equation (41) is more advantage than (40) and (40a). Results of the sieves analysis are given in table 3.

Using experimental data of granule formation the following values of coefficients is determined

$$\theta(L) = 3.0654\times10^{-7}L^4 - 5.401\times10^{-5}L^3 + 0.003292L^2 - 0.07553L + 1.3775$$

$$b(L) = -6.1205\times10^{-6}L^3 + 9.6722L^2 - 0.04745L + 0.80693$$

In **fig.5.** the calculation values (41) and (42) of evolution of distribution function and its comparison with experimental data of granule distribution on apparatus length are presented.

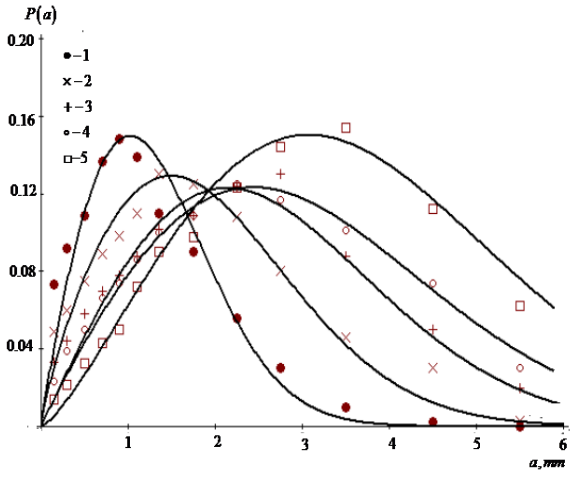

Fig.5. Particle size distribution in the granulation processes in drum. The solid lines represent predicted size distributions according to Eq. (41) for several distances from the powder entrance. the experimental points are for: 1– L=10 cm: 2– L=20cm: 3– L=40cm: 4–L=60cm; 5– L=75cm.

The distribution has a humped character as far as the probability of formation of two-humped distributions for cylindrical apparatuses is kept in the field of embryo formation, i.e. at $t < \tau_p$.

The dependence of granule strength on time of addition can be estimated using equation (30) as

$$\frac{d\ln\zeta}{dt}=\frac{1}{\zeta}\frac{d\zeta}{dt}=k$$

$$t=0,\ \Delta=\Delta_0$$

where $\zeta=\Delta-\Delta_\infty$ – absolute granule strength, $\Delta_0,\Delta,\Delta_\infty$ – initial current and limits value of strength, k – experimental coefficient. The solution of this equation taking into account of experimental investigations can be presented as $\left(\Delta_\infty >> \Delta_0\right)$

$$\Delta=\Delta_\infty\left(1-\exp\left(-kt\right)\right),\ \ \Delta_\infty\approx 0.37a-0.66$$

In **fig.6** the comparison of dependence of strength and yields of granule on time of addition are presented.

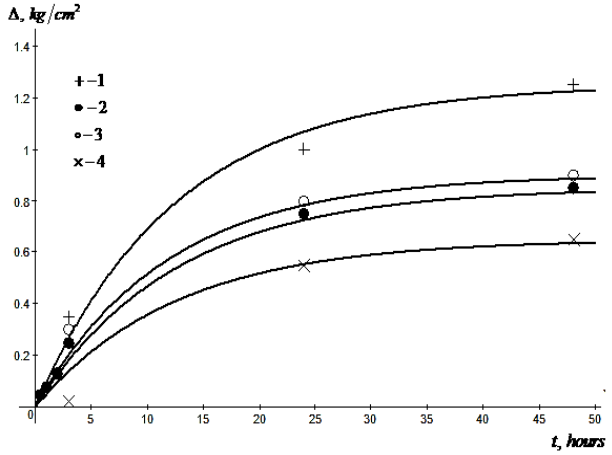

Fig 6. Changing strength titan-magnetite granules according to size and time:

1– $a=3.5mm$; 2– $a=4.0mm$; 3 – $a=4.5mm$; 4 – $a=5.0mm$.

In conclusion we note that the granulation process of titan-magnetite powdered materials is stochastic as far as the prepared granulemetric composition is polydisperse which is determined by uniform completeness of granules depending on drops sizes of binding substance (dispersion character), particles of powder and such phenomena as coagulation and destruction, wear and deformation. At the same time as a result of drum rotation the forming nuclei can also coagulate, which influences on the end granule size. Apparently, with increase of drops size of liquid and nucleus sizes, the probability of formation of granules of large size and large lumps is increased.

Notation

a – granule diameter, mm;
C_d – addition concentration, %;
C – concentration of binding substance;
D_* – efficient diffusion coefficient;
D_K – drops diameter, m;
L – length of cylindrical apparatus, m;
R – granule radius, mm;
R_P – pore radius, mm;
S – granule surface area, m^2;
V – linear velocity of granule mixing, m sec^{-1};
V_s – specific volume of porous medium, m^3 kg^{-1};
$\overline{U}_S$ – vector of making ones of velocity;
Q – yield of goods production, %;

Greek letters:

a – change rate of granule volume, m^3 sec^{-1};
ε – granule porosity;
Δ – granule strength, kg cm^{-2};
λ – laminating thickness, мм;
σ_D – deforming stress, H m^{-2};
ρ_p – poured density of layer, kg m^{-3};
ρ_d – density of particles, kg m^{-3};
η_S – volume viscosity of porous medium, H sec m^{-2};
ξ_S – shift viscosity of porous medium, H sec m^{-2};
τ_P – relaxation time of embryo formation, sec.

Compliance with ethical standards

Conflict of Interest The authors declare that they have no conflict of interest.

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