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## NANOCOMPOSITE ORGANOMINERAL HYBRID MATERIALS.

### PART III

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#### EXTENDED ABSTRACT:

The paper addresses the issues of alkoxide method of sol-gel synthesis and non-hydrolytic method of sol-gel synthesis and colloidal method of sol-gel synthesis. The authors also consider an alternative approach based on the use of soluble silicates as precursors in the sol-gel technology, of nanocomposites. It was shown that nanocomposites can be produced through aerogels. The paper also analyzes the mixing technologies of nanocomposites preparation. It has been demonstrated the possibility to change the types of nano-phase which is used for obtaining nanocomposites in different approaches. Various models of packaging spherical, fibrous and layered nanoparticles, introduced into the structure of the nanocomposite, in the preparation thereof were examined.

**Key words:** Nanocomposites, sol-gel synthesis, soluble silicates, metal alkoxide, sols, gels, aerogels, packing of spherical nanoparticles, packing of fibrous nanoparticles.

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## 4. INFLUENCE OF VARIOUS FACTORS ON STRUCTURE AND PROPERTIES OF HYBRID MATERIALS

### 4.1. Packing of spherical nanoparticles of the filler

The system of pores in a solid is entirely determined by the nature of the packing of the primary particles. For example, when the hexagonal closest packing of spherical particles between disposed in a plane, the three nearest tetrahedral cavities is octahedral cavity. In subsequent accretions over the octahedral cavity occurs again octahedral cavity. Superimposed on top of each other octahedral cavities form channels that permeate the whole body of matter in the orthogonal directions [151]. In cubic structures of the closest packing may also indicate the formation of interconnected channels. In the package of this type the void volume is 25.95%. From here it is possible to calculate the bulk density of the material ( $\rho$ ):

$$\rho = \rho_k (1 - L),$$

where:  $\rho_k$  – crystallographic density of the material used as a basis.  
 $L$  – void fraction in the structure of the package.



If the lightest of refractory materials has crystallographic density close to  $3 \text{ g/cm}^3$ , the bulk density of the product with the densest packing is  $2.2 \text{ g/cm}^3$ . From here, to obtain a materials with a bulk density less than  $0.5 \text{ g/cm}^3$ , it is necessary to achieve a greater porosity of 83%.

Let's try to analyze what kind of particle packing can achieve the desired porosity.

Typically with decrease in the coordination of number of particles packed in the packaging structures of particles increases the pore volume. In the case of the densest packings, the coordination number of the package is 12. Consider the following options for the coordination numbers of the package. Classically, such a packing density was 74%, respectively; the porosity of such package is 26%. For monodisperse materials denser packaging does not happen.

The coordination number is 8. Cubic volume-centered packing. In contrast to the dense packing it has a loose structure. Here the balls in a flat layer, not touching each other, and are at a distance  $0,155R$ . Motive construction of this structure is characterized in that the balls of the second layer are omitted in the voids of the underlying layer. Third layer repeats the first and the fourth – the second, etc. This type of packaging is a optimal variant spatial arrangement of spheres with a coordination number of 8, and has a lower percentage of its filling volume. The pore volume is 31.98%.

The coordination number is 6. Simple cubic packing. Globular structure, with a coordination number of 6, are built by successive overlay balls packed in a square grid so that the centers of the balls of the second layer are located directly above the centers of the balls first, third – above the centers of the second, etc. Pores in this package, enclosed between eight balls in contact, and has six throats, close in shape to a quadrangular. In this case formed is looser structure in which the pore volume is 47.64%

The coordination number is 4. Packing type of diamond and ice. The customized packaging is the most loose of all considered regular packages. It is constructed from layers of balls of spaced from each other in a plane. Balls in the next layer are between the balls of the first layer. Centers of the balls of the third layer are located directly above the centers of the spheres of the second layer. The fourth layer is located exactly above the first, etc. As a result of such packing of balls, the volume of voids produced is 65.99%. In the limit, with a decrease in the coordination number, each

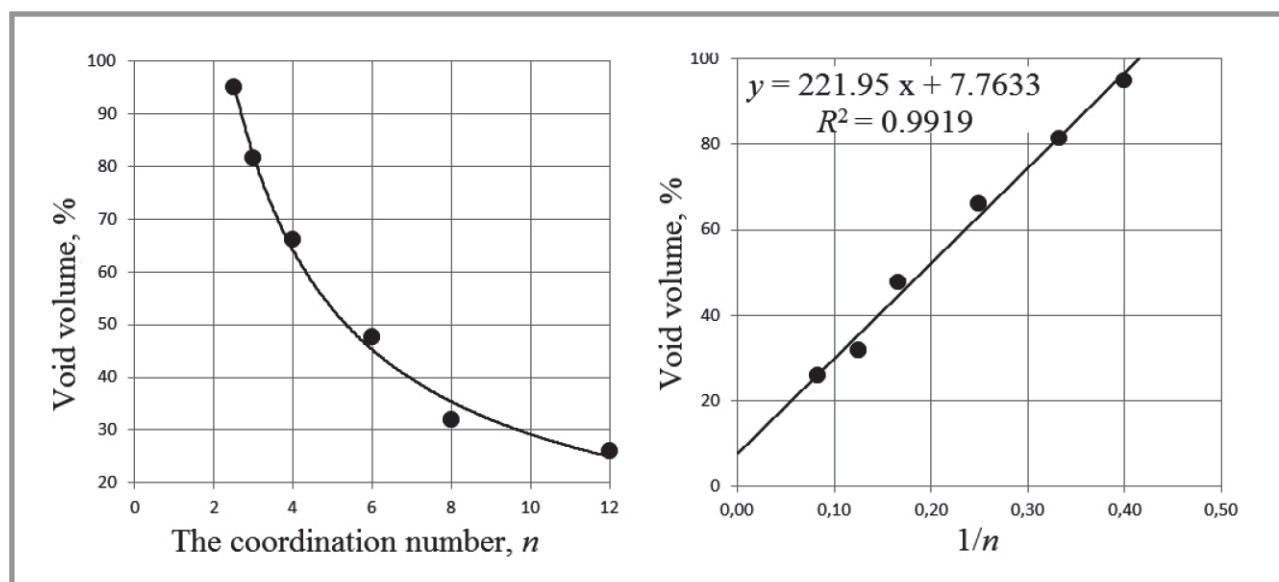


symmetrically constructed structure with a predetermined pore system should go into a chaotic folded body with a set of pores which will differ in both shape and size. Boundary of such a transition can be considered packing with a coordination number of 3.

The coordination number is 3. This package is a very fishnet, loose formations with a continuous three-dimensional framework. The porosity of this structure is 81.5%. Thus, relying solely on the various kinds of packing of spherical particles, it is not possible to achieve the desired porosity, and, moreover, to achieve the density less than  $0.2 \text{ g/cm}^3$ . However, if we admit the formation of mixed packing with a coordination number of 2 and 3, i.e., when some particles in the considered package present in the form of short chains consisting of at least three particles. For such packages can be achieved porosity – 95%.

The dependence of the porosity from the coordination number, for different methods of stacking spheres is shown in Fig. 16, which shows that the porosity is inversely proportional to the magnitude of the coordination number.

Currently particularly among the various sorbents is quite common such material in which a large part of their volume falls on the pores. For example, the porosity of silica gels ranges from 36 to 84% [153]. In [154]



**Fig. 16.** Dependence of the porosity of the packing of spherical particles from the coordination number  $n$  and  $1/n$ .



describes the preparation of aerogel with a porosity of 99%. Aerosils porosity is 95÷98% [155]. Similar structures are formed from fibrous crystalline silicates such as chrysotile, during their disintegration by a surfactant. Thus individual fibers of the tubular type [156] with a diameter of 18 microns are formed, that due to the weave creates very loose stable colloidal dispersions with a high porosity [157]. Apparently, such net structures have liogels, aerosils and some, particularly loosely-packed xerogels, globular chains which are interconnected so that the number of contacts for the majority of the globules is 2. For stiffening the skeleton of the gel, the number of contacts between the particles and their neighbors, at the nodes of this network should be equal to 3. But in any case such structures constructed on the basis of the spherical particles do not have sufficient rigidity. Therefore, the most promising methods for the preparation of porous materials having sufficient strength is the use of different fibers or hollow microspheres.

Based on the plot of the porosity  $\alpha$  (%) from the coordination number  $n$  of spherical particles, shown in Fig. 16, one can obtain the following dependence:

$$\alpha = \alpha_{\infty} + \frac{B}{n}.$$

The coefficients in this equation for the dimension  $dim(\alpha) = \%$ , have the following meanings:  $\alpha_{\infty} = 7.763 \pm 0.016$ ;  $B = 221.95 \pm 0.45$ ,  $R^2 = 0.9959$ . Since the equation of this curve has a singular point, it is possible to calculate the limiting value of the coordination number, in which the void volume will be equal to 100%:

$$n_{100} = \frac{100 - \alpha_{\infty}}{B}.$$

Value  $n_{100} = 2.406 \pm 0.010$ . The physical meaning of this value is that in dispersed nanostructures with coordination number less than this value is missing more or less stiff the carcass structure of the material collected from individual nanoparticles. Below, this is the coordination number; the particles are collected only in the individual chain structures that are not related to each other in which is realized coordination number 2.

To describe the dependence of porosity on the coordination number of the packing of spherical particles, calculations were carried out for the se-



lection of correlation, which would give a maximum correlation coefficient. The maximum correlation coefficient was obtained for the dependence of the following form:

$$\alpha = \alpha'_\infty + \frac{B'}{(n - n_0)}$$

The coefficients in this equation, under conditions as in the previous case were:  $\alpha'_\infty = 0.14964 \pm 0.00019$ ,  $B' = 310.10 \pm 0.40$ ,  $n_0 = -0.7750 \pm 0.0010$ ,  $R^2 = 0.9975$ . The resulting correlation dependence is shown in Fig. 17. For this relationship limit value of the coordination number at which the void volume is equal to 100% can be also calculated:

$$n_{100} = n_0 + \frac{B'}{(100 - \alpha'_\infty)}$$

Value  $n_{100} = 2.331 \pm 0.006$ . To interpret these results is more difficult, even though they give a more accurate result, and several of them are expanding the range of coordination numbers, to which still may form rigid skeleton, with openwork packing of spherical particles.

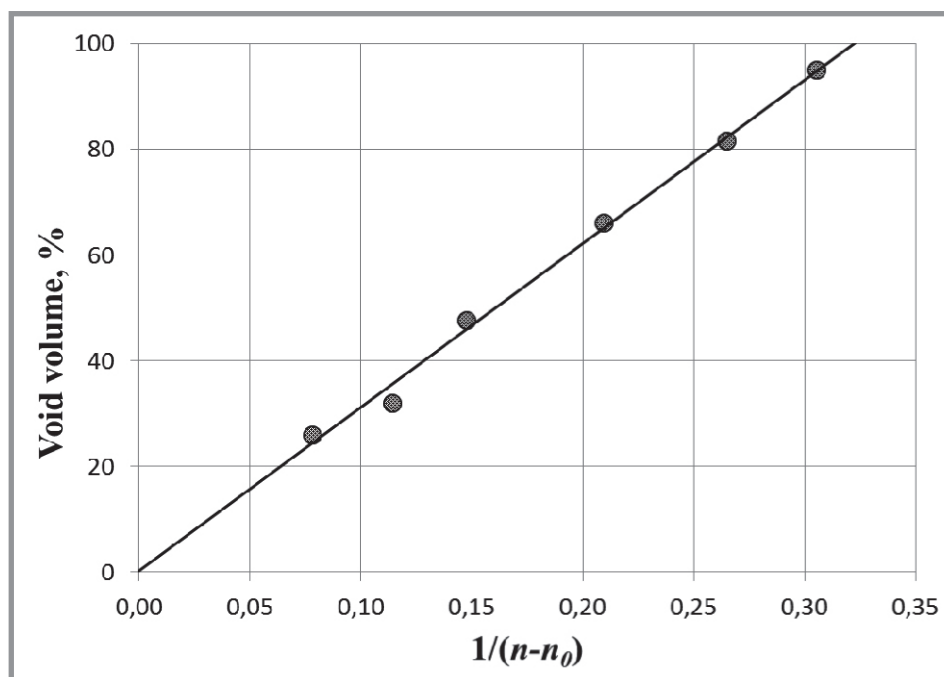


Fig. 17. Dependence of the porosity of the packing of spherical particles from  $1/(n-n_0)$ .

In recent decades the rapid development of computer technology has led to the creation of fundamentally a new powerful means of investigating the physical processes – computational experiment. Numerical simulation is the only way to study these processes in conditions when theoretical methods meet with serious difficulties, and practical experiments or overly laborious, or fundamentally impossible. Creation of computer models has now become a recognized and rapidly developing direction of science and technology. Application of simulation compared to traditional laboratory methods saves money and time. In studies based on computational experiments, the crucial role played by the representation of the physical phenomenon in the form of an appropriate mathematical model, computational algorithm, a computer program. Mathematical model in the computational experiment is simultaneously a test material and the algorithm of the experiment. In [158] it is shown that one of the most common methods of constructing mathematical models is the simulation by means of particles or particle method.

It is also indicated that, when used properly particle method is able to demonstrate its clear advantages. On the basis of the accumulated theoretical and experimental data [158–160] a computer model of disperse systems based on the particle method was built.

Particles method is common to the class of models in which discrete description of physical phenomena, including consideration of the interacting particles. Any classical system consisted of particles can be described, if one knows the law of their interaction, their coordinates and velocities. Each particle has a number of fixed characteristics (e.g., weight and size) and changing characteristics (eg, position and velocity). As an object of numerical study of disperse systems by computer simulation was chosen aluminum powder. This is due to the fact that the aluminum particles have a spherical shape. As a result of these calculations were obtained varying degrees of, the volumetric filling of system under the external pressing force. From the presented in Fig. 18 data on the distribution of the coordination numbers for different volumetric filling of the powder follows that the limit volumetric filling of powder equal to 64% corresponds to a state where more than 40% of the particles has a coordination number of 12.

The computer model also makes it possible to study the nature of the contacts between the particles in the dispersion. To investigate the distance between «contacting particles» corresponding computational experiments



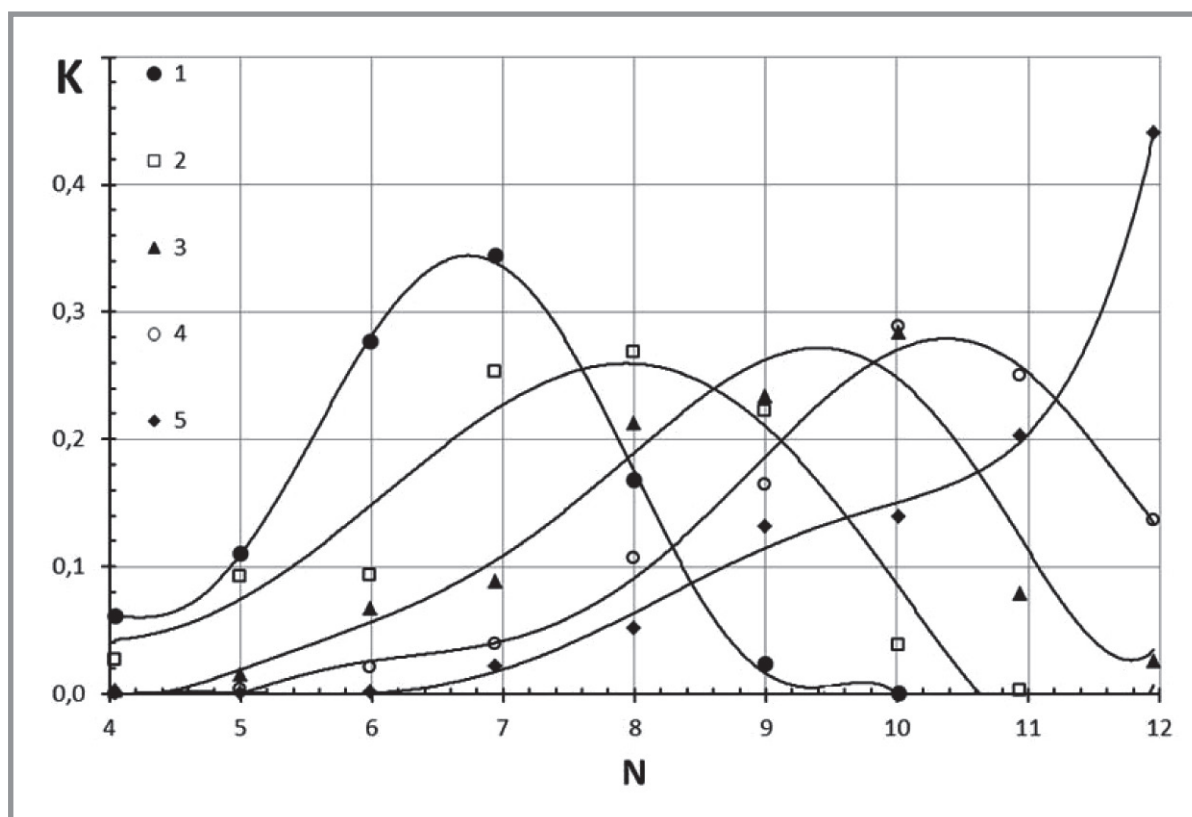
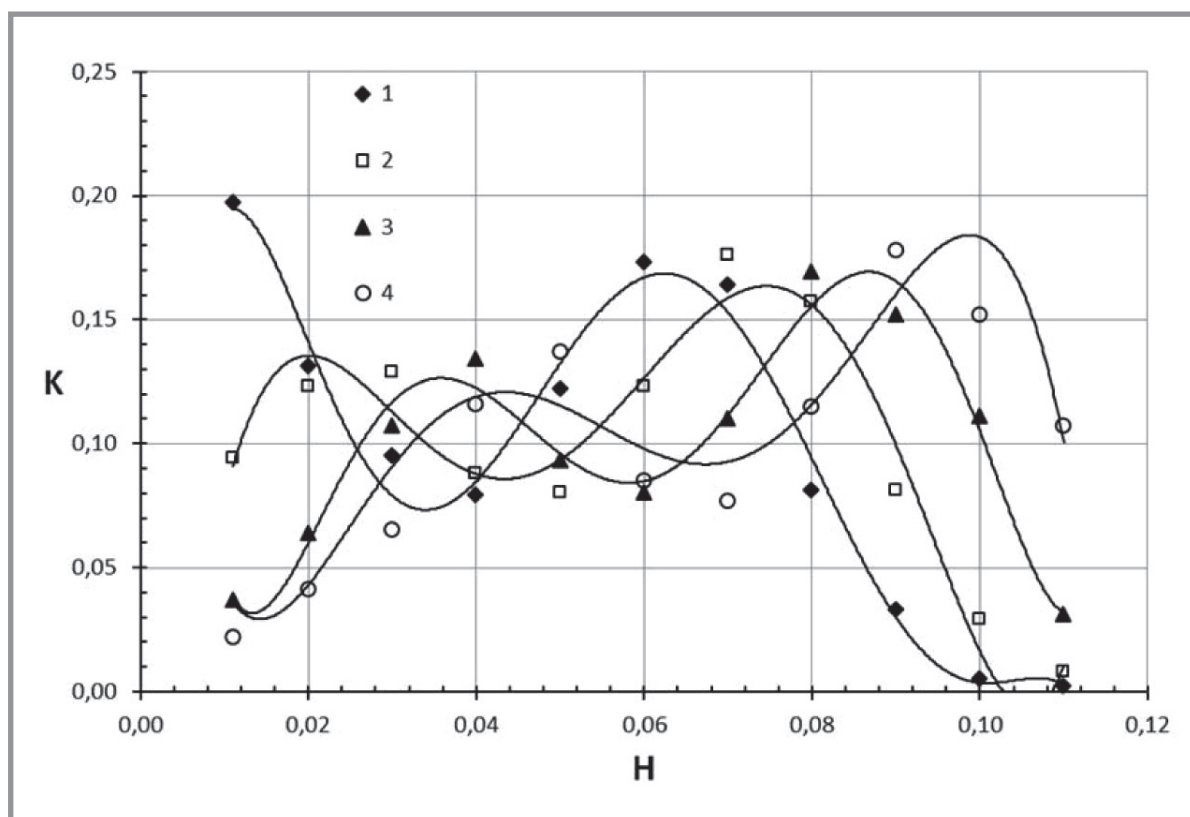


Fig. 18. Distribution of coordination numbers for different volumetric filling of powder: 1 – 0.56; 2 – 0.58; 3 – 0.60; 4 – 0.62; 5 – 0.64.

were conducted. Fig. 19 shows the distribution of the number of pair contacts depending on the distance between the particles for different volumetric filling. From this figure, it follows that for all volumetric filling dependence of the distribution of pair contacts between the particles of the distance between them has two peaks. Reducing the volume filling of the system leads to a shift of the maxima in the region of the greater distance between the particles.

Storage and processing of a powder in real conditions is accompanied by absorption of moisture from the surrounding medium as a result of water vapor adsorption on the particle surface and its capillary condensation in the pores of the material. The formation of the liquid phase leads to the appearance of liquid «bridges» and capillary forces between the particles of the powdered material. These forces have a significant impact on the behavior and properties of the powders and dispersion compositions, both in storage and when using in industrial processes. In this case the various pa-





**Fig. 19. Distribution of the pair of contacts of the particles as a function of the distance between them for different volumetric filling of system: 1 – 0.64; 2 – 0.62; 3 – 0.60; 4 – 0.56.**

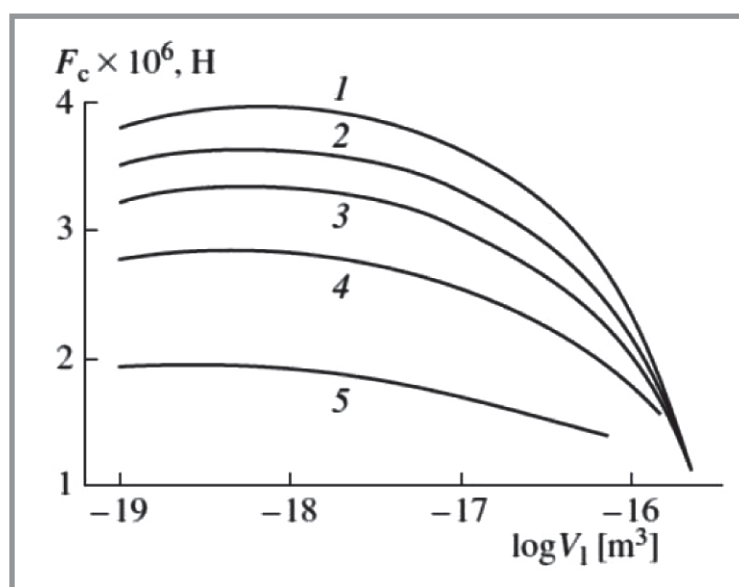
rameters of the particles have a significant impact on the amount of capillary forces, in particular, the size and shape of the liquid «bridges» causing a type of particle «contacts». Consequently the determination of the effect of geometrical parameters of the powder particles, by an amount of the capillary forces acting between them, allow us to predict better the properties and behavior of the particulate components and will also contribute to obtaining, based on them, disperse materials with specified properties.

The proposed method also allows determining the amount of liquid in a capillary «bridges» between the particles, and the magnitude of the capillary pressure as a parameter, which determines one of the components of capillary forces [161]. As the computational cell, has been considered capillary interaction of two particles connected by a liquid «bridge», and having the shape of a sphere, a cone and a plane. In real disperse systems using the selected forms can be described a large number of various types of «contact» between the particles.

In this case, as in the case of spherical particles of the same size, the magnitude of capillary forces it is dependent on two components. The first component is determined by the surface tension of the liquid, which acts along the wetted perimeter. The second component is caused by the presence, rarefaction or pressure arising due to the curvature of the surface of the «bridge» liquid, and is described by the Laplace equation.

It is seen that in all cases, growth of quantity of the liquid accompanied by the appearance maximum of capillary forces. Growth of the capillary force occurs in accordance with the increase of the ratio of particle sizes (see Fig. 20), the magnitude of this force is committed to the values corresponding the type of «contact» particle, such as a «sphere-plane». This effect is particularly noticeable with large volumes of liquid «bridge» when the differences in the values of force are negligible. Based on the obtained data it was not only qualitatively confirmed and quantitatively shows the influence of differences in particle size by an amount of capillary forces acting between them.

It was determined that the use of these equations depends on capillary forces, on the value of the amount of liquid in the «bridge» for the considered types of «contact» of the particles (see Fig. 21). Water is considered



**Fig. 20. The dependence of the capillary force of the volume of liquid «bridge» in the interaction of particles of type «sphere-sphere». Radius of the particle is equal to 5 microns. Ratio of the radii of the particles is equal to: 1 – 100, 2 – 10, 3 – 5, 4 – 2.5, 5 – 1.**

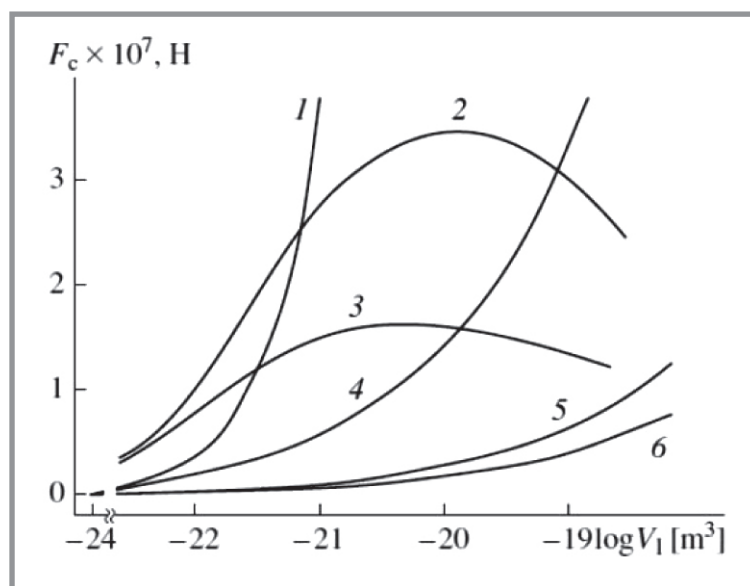


Fig. 21. The dependence of the capillary force of the volume of liquid «bridge» for any type of «contact» between the particles:

- 1 – «plane-plane» ( $d = 0.2 \text{ mkm}$ ), 2 – «sphere-plane» ( $R = 0.5 \text{ mkm}$ )  
 3 – «sphere-sphere» ( $R_1 = R_2 = 0.5 \text{ mkm}$ ), 4 – «cone-cone» ( $\beta_1 = \beta_2 = 150^\circ$ ),  
 5 – «cone-plane» ( $\beta = 90^\circ$ ), 6 – «cone-cone» ( $\beta_1 = \beta_2 = 90^\circ$ ).

as a capillary liquid. For all types of «contact», the size of the gap between the particles is equal to  $0.01 \text{ mkm}$ , the wetting angle was  $0^\circ$ . Fig. 21 shows that type of «contact» of particles has a significant effect on the magnitude of the capillary force. Its influence increases with increasing volume of «bridge».

On the basis of the proposed models the rheological properties of organo-mineral nanodispersions and the possibility of obtaining on their basis the conductive polymer composites experimentally have been described and tested [162, 163].

#### 4.2. Packing of fibrous nanoparticles of the filler

Heterogeneous structure is one of the most important factors determining the mechanical behavior of reinforced, dispersion-strengthened nanocomposites under mechanical loads. Currently, there is a need for modeling spatial patterns of nanocomposites in order to predict their effective elastic and strength characteristics. To exclude the possibility of obtaining a weakly predictable results, which is especially important for

the problems associated with the generation of materials with high volume fractions of fibers it is necessary to use specially constructed iterative procedures to streamline the reinforcing elements.

Synthesis of fragments of random structure of fibrous nanocomposites, the reinforcing elements, which are fiber, round in cross-section, is related to the random placement of disjoint smooth disks in the plane. In [164, 165] the various ways of generating of fibrous and dispersion-strengthened materials are described in detail. To determine the laws governing the formation, rarefied and close-packed structures, of reinforced nanocomposites, and estimation of influence of factors that affects their properties, has implemented the following algorithms.

Algorithm 1 is Monte Carlo method. In the studied fragment (typically a single square) is randomly placed fibers. If, in the generation of fiber position, it is not beyond the fragment then coordinates of the center of the fiber are recorded. In the synthesis of the following coordinates of fibers, checks crossing fibers placed previously. In the case of both of the two conditions: the absence of intersections with all reinforcing elements and the location in the fragment generated the fiber is «turned on» in the structure of the composite. Upon reaching the necessary or highest possible volume fraction of filling, or to fulfillment of the condition, when limit value is exceeded number of unsuccessful attempts to accommodate reinforcing elements.

Algorithm 2 is an additional displacement fibers at the «hard» boundaries of the fragment. This algorithm is a modification of the method of «radial gravitational field», features a software implementation, which are described in detail in [164]. Simulating a random structure is carried out additional mutual movement along a straight line connecting the centers of the cross-sections, again, and the previously generated fibers, at a distance, which guarantees the absence of intersections with all the structural elements. For any part of cross-section in the reinforcing element the exceeding of the domain bounds is completely excluded. If during the placement correction some fiber comes into contact with the boundary of the fragment, its movement may be only along this border. Fiber simulation and modification of their location are made to create a random structure with the given volumetric filling, or reaching a certain limiting value the amount of additional relocatable fibers.

An important characteristic of random structure of composites is the volume fraction of the reinforcing filler. For the definition of limit of the



volume fraction fibers, it is necessary to choose parameter that is the criterion that determines the end of the iteration process. As the criterion for algorithm 1 the total number of unsuccessful attempts fiber placement can be used. For Algorithm 2, such criterion is the relative number of additional relocatable fibers (the normalization is carried out to the total number of fibers) in the simulated structure of the composite. In this model there is a parameter that determines the guaranteed thickness of the layer of the matrix  $d$ , surrounding each fiber, which is characteristic of the inter-particle interaction. This parameter is given in the form of relative  $d / R$  ( $R$  – radius of the fiber). An output dependency of the volume fraction from these parameters (see Fig. 22) on the horizontal asymptote corresponds to the moment they reach the maximum volume filling.

Simulation was performed with composites formed by circular in cross section fibers of the same diameter, surrounded by layers of the matrix material of different thicknesses. Table 2 presents data on the limiting volume fractions of filling obtained by averaging of 20 independent realizations of the random structure. Size of guaranteed interlayer of the matrix is a parameter that depends on the nature of the components of the nanocom-

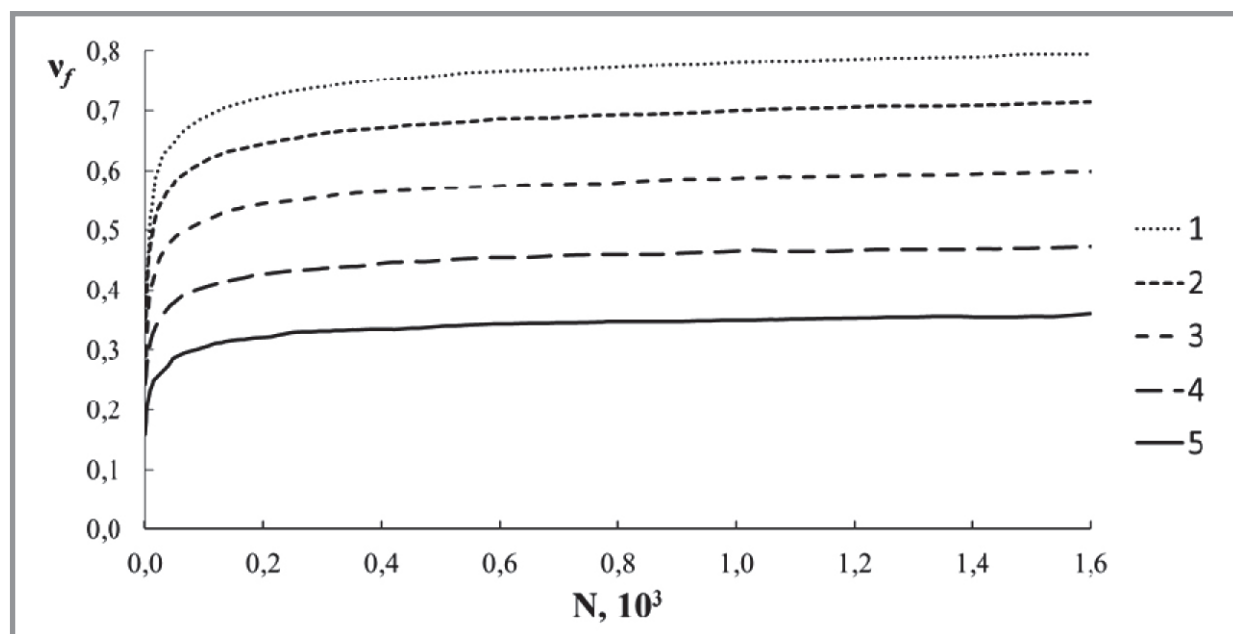


Fig. 22. Dependence of the volumetric filling of the composite on the relative number of fibers, relocatable further. Thickness, of guaranteed, layer of the matrix  $d/R$  is: 1 – 0,0; 2 – 0,1; 3 – 0,3; 4 – 0,6 и 5 – 1,0 [165].

Table 2.

**Limiting the volume fractions  $v_f^{\max}$  circular in cross section fibers**

The guaranteed thickness of the layer of the matrix $d/R$	0.00	0.10	0.20	0.30	0.40	0.50	0.60	0.70	0.80	0.90	1.00
Algorithm 1	0.53	0.48	0.44	0.40	0.37	0.34	0.31	0.29	0.27	0.25	0.24
Algorithm 2	0.80	0.73	0.66	0.61	0.55	0.51	0.48	0.45	0.42	0.40	0.37

posite, the technology of the material composition and properties of special coatings and adhesive layers applied to the reinforcing filler in the manufacture of the composite, the parameters of the electric double layer or the nature of the van der Waals forces. As can be seen, increasing the thickness guaranteed, interlayers, to a size corresponding to the radius of the fiber, leads to a reduction limit of the volume fraction more than doubled.

In real composites the volume fraction of the reinforcing filler is typically 0,60÷0,70, and in highly materials – 0.80÷0.85. To obtain fragments of random structure with the volume, degree of filling, exceeding  $v_f^{\max} = 0.50$ , was used by Algorithm 2, in which, provides an iterative procedure, the additional displacement of the fibers. It was found that the value  $v_f^{\max} = 0.80$  (same cross-sectional fiber without guaranteed layer) exceeds the highest possible degree of filling of the composite with a periodic arrangement of fibers in the tetragonal grid nodes ( $v_f^{\max} = 0,785$ ). However, it does not reach the limit of the volume fraction, typical of the material with a periodic hexagonal structure ( $v_f^{\max} = 0,907$ ). Recorded values  $v_f^{\max}$  coincides with the value of the maximum filling, for random dense packings of «smooth» disjoint disks of the same diameter, obtained by the authors [166].

These papers presented data related to the materials with a high filling, which is particularly important for structural materials. However nanocomposites are also important low degree of filling of the fibers. This calculation was presented in [167].

Comparison of the packing of spheres described in the previous section and the fibers can be represented by the following analogy between open-



work packing of spherical particles and fiber packing. If the chain of bonded spheres represented in the form as described around them, cylinders, in this structure it is possible to build a relationship between the average coordination number of spheres and the ratio of fiber diameter  $d_v$  and the average distance between the points of contact between two adjacent unidirectional fibers intersecting and touch the third fiber  $l_{av}$ . For  $n = 3$ ,  $l_{av} = 2r_s = d_v$ , where  $d_v$  – fiber diameter, and for  $n = 2$ ,  $l_{av} = \infty$ .

Character of packaging of fibrous particles can be determined by the structural parameter that has the form:

$$p = \frac{l_{av}}{d_v}.$$

Tracing the analogy of packing fiber and spherical particles, the effective coordination number of the packing can be represented as follows:

$$n = c + \frac{1}{p},$$

where  $c$  is a constant quantity.

From the equation of porosity, in Section 4.1., we obtain:

$$\alpha = \alpha_{\infty} + B \frac{p}{c \cdot p + 1},$$

$p \rightarrow \infty$ , porosity of  $\alpha \rightarrow 100\%$ . From this we can estimate the value of the constant  $c$  (when  $\alpha_{\infty} = 7.763 \pm 0.016$ ;  $B = 221.95 \pm 0.45$ ):

$$c = \frac{B}{100 - \alpha_{\infty}} = 2,406 \pm 0,005 .$$

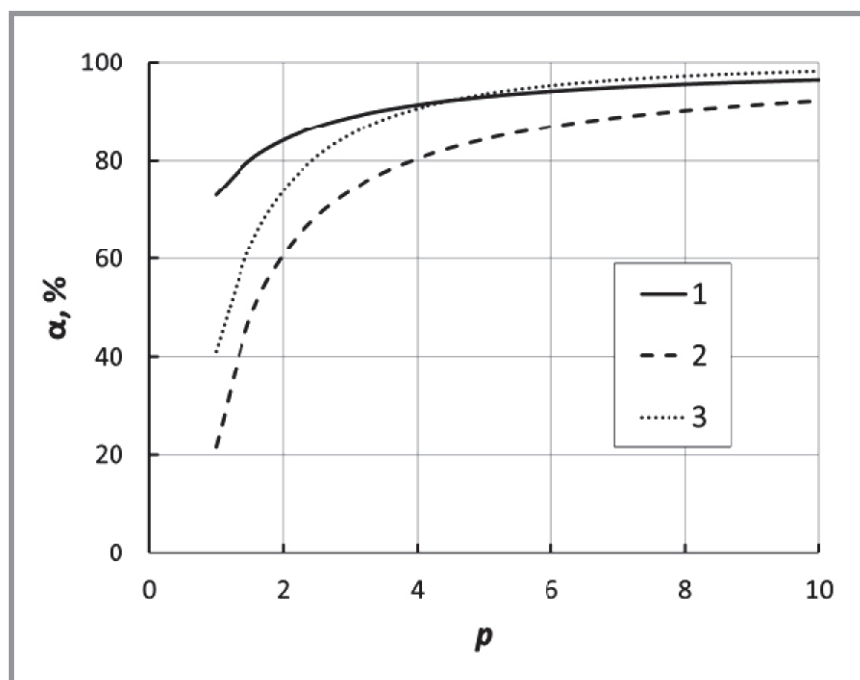
The given geometry package has an openwork character. Graphically equation such packing of fibrous particles is shown in Fig. 23, curve 1.

To verify this model we will consider other models of geometric packing of fibrous particles. The first model may be represented as alternating, mutually orthogonal layers of endless fibers. It is easy to see that the porosity in this model would have the form:

$$\alpha = 100 \left( 1 - \frac{\pi}{4 \cdot p} \right).$$

Graphically this dependence is presented in Fig. 23, curve 2.





*Fig. 23. The dependence of porosity ( $\alpha$ , %) from the structural parameter  $p$  fibrous packing of nanoparticles: 1 – packing openwork fibers; 2 – interspersed layers of fibers are mutually perpendicular; 3 – arranged regularly in space simplexes of the three mutually orthogonal fibers.*

The second model is located in the space regularly, simplexes of three mutually orthogonal fibers. For this model we can easily show the dependence of porosity on structural parameter:

$$\alpha = 100 \left( 1 - \frac{3 \cdot \pi}{4 \cdot (p + 1)^2} \right).$$

Graphically this dependence is presented in Fig. 23, curve 3. Comparing the dependence we can see that the densest packing is mutually orthogonal layers of fibers. Openwork packaging and packing regular simplexes of three mutually orthogonal fibers are closest to each other, especially for  $p > 4$ . This effect is typical for the most rarefied structure. Different behaviors of dependencies, for small values of the structural parameter, due to the fact that the model geometry openwork provides packaging of discrete fibers and other models consider packing continuous, endless fibers. Thus, we can assume that the curves obtained in the framework of openwork packaging are close enough to realistically obtain the package.



### 4.3. Nanomaterials based on layered particles

Currently these composites are widely used in industry, both as structural materials and in other uses. Addition of them to the polymer, even small amounts of Silicate nanoplates (usually 3–5 wt. %), can significantly improve the barrier diffusing material properties, thermal stability, resistance to thermal warping [168–172]. This is due to the fact that in contrast to conventional composites (single components of conventional composites have micron and submicron sizes) to nanomaterials characterized extremely high interfacial area boundaries, resulting in their role in the formation of physical properties of the material becomes dominant. It is possible to obtain significant gains in improving different physical characteristics of nanomaterials with very low concentrations of filler. In the nanocomposites the volume fraction of the particles is usually only a few percent, whereas in conventional composites is on the order above [173].

For the first time the use of ultrafine clay filler was proposed in 1974 [174], but only recently great demand for the application of such materials are really in great demand. Currently one of the most common types of nanocomposite is the systems based on polyolefins and layered clay minerals (smectite). In the past decade the world experienced a rapid growth in the number of publications and patents related to the manufacturing process and the study of their mechanical properties [171, 175, 177]. Such systems achieved a substantial increase in the elastic modulus, strength, fire resistance, resistance to thermal warpage improve barrier properties with respect to the diffusing substances. However, analysis of well-known publications shows that the industry of materials in terms of understanding and explanation of available evidence, data, designed, is still not enough.

For relevant research, as a binder matrix, widespread polymer such as polyethylene was used. This is partially crystallized material, and even in its pure form is a complex multi-level hierarchical structure that is a well-defined structural heterogeneity on the nano-, meso- and micro levels [175].

These systems are structurally inhomogeneous medium that consists of polyolefin matrix and implanted in it ultra-thin silicate flakes, which thickness is a few nanometers, and average diameter from tens of nanometers to 1  $\mu\text{m}$ , depending on the mineral deposit and the conditions of its formation. Typical filler particle sizes are 1–2  $\text{nm}$  in thickness, and from



30 to 1000 *nm* in diameter. These particles can be chaotically distributed in the volume of material or can form individual packs – tactoids. Tactoids consist of several (usually on the order of tens) of parallel plates, between which one or more molecules of the polymer matrix [172] are located. In the first case the nanocomposites are called exfoliated materials, in the second – intercalated materials.

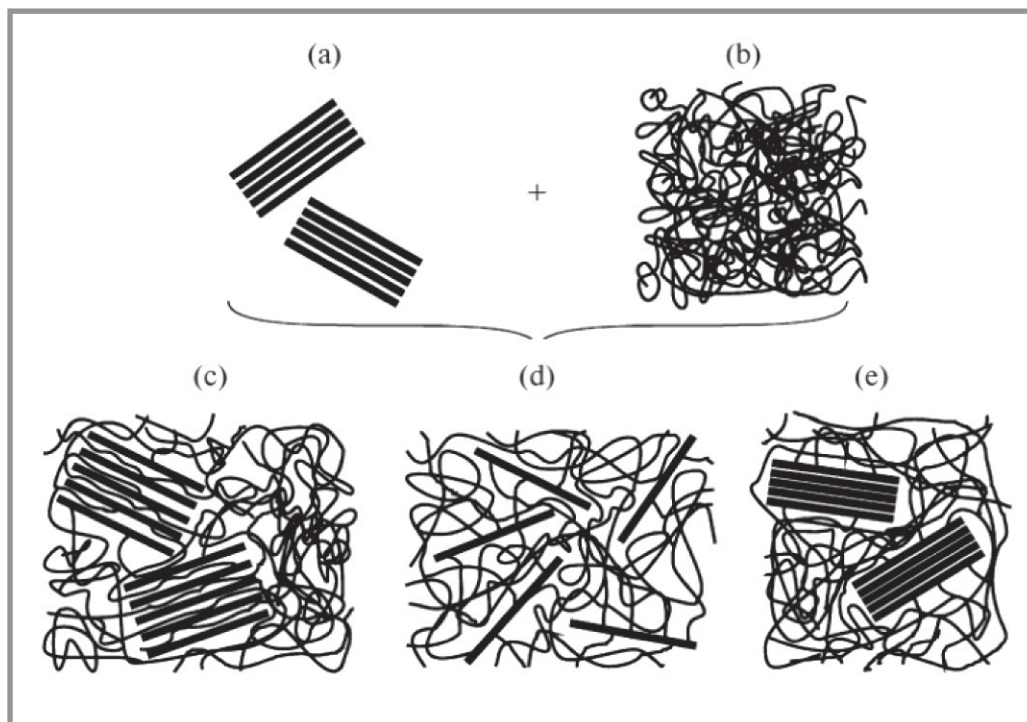
At low thermodynamic compatibility of the filler and the matrix polymer molecules cannot penetrate into the gaps between the layers of the silicate. The result, in this case, is that the material is a simple mechanical mixture of polymer and mineral. This is the usual, dispersion-filled, microcomposite [178]. When forming intercalated nanocomposite polymer molecules diffuse into the space between the closely spaced parallel plates of silicate. As a result there is a swelling of the mineral crystallite, but in this case there is no loss order in the arrangement of its layers. In the formation of exfoliated nanocomposite polymer molecules penetrate into the gaps between the layers of tactoids, and move apart them so dramatically that there is a complete destruction of the tactoids, and silicate plates previously parallel acquire a random orientation [69, 179].

When formed exfoliated nanocomposite, polymer molecules not only penetrate into the space between the layers of tactoids but they also force them to interact with each other. Thus, the structure collapses, and at the same time the silicate plates lose its orientation in space and are randomly distributed in the bulk material. Figure 24 is a schematic representation of the structure of the nanocomposite in cases intercalated and exfoliated filler [171].

Mechanical properties of the nanocomposite improve if the silicate filler particles are oriented along the direction of the external deformation. This occurs due to improved adhesion between the matrix and the inclusions due to «biting» of the matrix and also when the outer plates are bent inwards and on the internal plates are absent bends. This effect can be achieved by pretreatment of the material, for example, by drawing through a die or other similar methods.

The concentration of the filler to a greater extent affects the increase of the macroscopic elastic modulus. When the amount of the filler is only 6–8%, its elastic modulus increases approximately by 3 times. If we compare composites with granular filler, the famous Einstein's formula gives an increase in module only 1.2 times [180].





*Fig. 24.* Schematic representation of the different structural types of nanocomposites polymer/clay: (a) crystallite clay (tactoid), (b) base polymer, (c) intercalated nanocomposite, (d) an exfoliated nanocomposite, (e) phase separation nanocomposite.

## CONCLUSION

Synthesis and study of the properties of new organo-mineral hybrid nanocomposites is of great importance. On the basis of these studies solutions to many specific technological problems can be found. Promising areas in the field of organo-mineral hybrid nanocomposite materials will be methods of synthesis of multicomponent materials, as well as materials such as «net in net» and «host-guest». The application of alkoxides of various metals, including complex, polynuclear and multi-functional in the sol-gel processes has a great potential. The fundamental problem with the chemistry and physics of nanocomposites remains the dependent «structure-property». Solution of this problem allows passing from research of materials to their purposeful designing. This task motivates various experts around the world to actively work on a solution.

**DEAR COLLEAGUES!**

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## НАНОКОМПОЗИТНЫЕ ОРГАНОМИНЕРАЛЬНЫЕ ГИБРИДНЫЕ МАТЕРИАЛЫ. ЧАСТЬ III

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### АННОТАЦИЯ К СТАТЬЕ (АВТОРСКОЕ РЕЗЮМЕ, РЕФЕРАТ):

В работе рассматриваются вопросы, связанные с алкоголятным и негидролитическим методами золь-гель синтеза, а также коллоидным методом золь-гель синтеза. Также для нанокompозитов обсуждается альтернативный подход, основывающийся на использовании растворимых силикатов в качестве продукта предшествующей стадии в золь-гель технологии. Показана возможность получения нанокompозитов из аэрогелей. Был выполнен анализ использования смешанных технологий при приготовлении нанокompозитов. Авторы описывают возможность изменения различных подходов в типах нанофаз, используемых для производства нанокompозитов. Исследованы различные модели упаковки сферических, волокнистых и слоистых наночастиц, введенных в структуру нанокompозита.

**Ключевые слова:** нанокompозиты, золь-гель синтез, растворимые силикаты, алкоголяты металлов, золь, гели, аэрогели, упаковка сферических наночастиц, упаковка волокнистых наночастиц.

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