NANOSTATE PHENOMENON IN MATERIALS SCIENCE OF METAL-POLYMERIC SYSTEMS

ФЕНОМЕН НАНОСОСТОЯНИЯ В МАТЕРИАЛОВЕДЕНИИ МЕТАЛЛОПОЛИМЕРНЫХ СИСТЕМ

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Abstract: *There were considered the physical, structural and morphological prerequisites for the realization of the nanostate phenomenon of dispersed particles of condensed matter of different composition, nature and technology for production. It was shown the role of the size factor in the occurrence of the nanostate phenomenon due to the change of the energy parameters of the surface layers of particles that contribute to their effective modifying effect on the high-molecular matrix. Physical models of the formation of a particular energy state of dispersed particles and metallic and non-metallic materials substrates, characterized by the presence of local areas ("charge-mosaic") with a long relaxation time are proposed.It was considered practical application of the nanostate phenomenon when creating high-strength and wear-resistant materials based on thermoplastic matrices (PA6, PTFE, PET), consistent lubricant and lubricating oils, tribological and protective coatings for friction units and metalwares used in mechanical engineering, automotive and mining engineering. It was made the examples of the effective use of developed nanocomposite materials in practice.*
KEYWORDS*: NANOSTATE, SIZE CRITERION, MODIFYING EFFECT, NANOCOMPOSITE MATERIALS, TRIBOTEHNICAL COATING, LUBRICANT.*

1. Introduction

Cutting-edge areas of functional materials science of engineering is the practical realization of the phenomenon nanostate at different levels of structural organization, providing synergistic effects increasing the parameters of strength, tribological, thermal, adhesive and protective characteristics of composites by using nanoscale components of different composition, structure and technology of [1- 10].

Despite the significant progress made in the composite nanomaterials, there are considerable difficulties in their industrial production and industry use, due to the absence of uniform evidence-based approaches to the theoretical justification of engineering technology of nanomaterials that determine directional component selection, methods for their production and processing to ensure their industrial production scoped application.

Certain aspects of the structural and morphological characteristics and electrical energy in the form of nano-dispersed particles, thin films are considered in studies of domestic and foreign scientific schools [3, 5-10]. However, the transformation of phenomenological phenomenon nanostate in engineering technology functional nanocomposites based on macromolecular (polymeric, oligomeric, mixed) matrices is only possible in the implementation of a systematic approach that takes into account the effect of the characteristic features of the structure, the morphology, the energy state of nanoscale components on the structural organization of macromolecular matrices at the molecular, supramolecular and phase levels.

The purpose of this study was to develop criteria for evaluation of physical nanostate dispersed particles of condensed matter to justify the engineering technology of nanocomposite materials based on polymeric, oligomeric and common thermoplastic matrices.

2. Methods of research

The main objects of the study were chosen nanoparticulate carbonaceous particles (graphite, UDAG, shungite, carbon fibers), metal (oxides, salts of organic acids) and silicon (mica, tripoli, opal clay) the compounds obtained by the process effects on natural and synthetic semi-finished products produced by industrial enterprises of Belarus and the Russian Federation. Nano-sized particles were prepared by mechanical grinding and heat treatment of disperse semis at temperatures 673-1473 K.

As matrix polymers used two basic types of materials. The first - with hereditary high viscosity (RGR) of the melt due to the chemical structure and molecular weight of the chain: polytetrafluoroethylene (PTFE), and ultra-high density polyethylene. The second group consisted of thermoplastic polymers PA 6, HDPE, EVA, PP, TPU, etc. With normal melt viscosity, this increases 2-3 times when the filling of nanoparticles - acquired high viscosity. Polymeric materials used in the state of industrial supplies as granules or powder produced by cryogenic dispersing granules at a temperature of 87 K.

The structure and properties of nanocomposite materials and their products investigated using modern methods of physical and chemical analysis: IR transmission and ATR (Specord), EPR spectroscopy (ER 1306, Brucer), X-ray diffraction (DRON 2.0, 3.0 DRON), differential thermal (Q-1500) analysis, optical (MIM-10, MF-2), scanning electron (ISM-50A, Nanolab-7) and atomic force microscopy (Nanotop III). Energy state nanomodifiers and composite materials was evaluated by EPR spectra and the spectra of thermally stimulated currents (TSC) on the original installation of the GNU MPRI them. VA White NASB. The dielectric characteristics of materials after exposure to energy (laser, ion, temperature) were determined by an appropriate standardized methods. Regulation nanorelief surface layer of polymer samples was performed by short-pulse laser and accelerated ion impact with a given power density. Evaluation features crystal-chemical structures of nanoparticles was performed by the original method, developed on the basis of X-ray analysis.

3. Results and discussion

To assess the role of the energy factor in mechanisms of formation of the structure at the molecular composite materials, and interface supramolecular structure parameters investigated dispersed particles, which are widely used as modifiers for oligomeric, polymeric matrices and combined.

Methods OM, AFM, SEM analysis of morphological features of dispersed particles of different composition, technology of reception, crystal chemical structure - oxide compounds metallurgical production (OM), the products obtained by a mechanical crushing silicate glasses (SS), carbon-fiber (CF), the products of detonation synthesis (UDAG), carbon nanotubes (CNTs), silicon-containing natural particles (CN) - mica, tripoli, clays, schungite, talc.

It was found that, regardless of the technological history and crystal structure of the starting semi-dispersed particles form the cluster structures, and consisting of single particles with nanoscale parameters. This single particle morphology characterized by the presence of nanoscale spherical elements, plate or whisker habit forming nanorelief surface layer, contributing to the formation of the boundary layer at the interfacial interactions of the components in the formation of the nanocomposite material. Specific nanorelief surface layer due to crystal-chemical and technological prerequisites, is predominant factor in the choice of the original semi-finished product and technology of dispersed particles with optimal dimensional parameters. This dispersion of particles of a modifier can be in the range of micro, providing the necessary modifying effect due to the presence of nano relief with a characteristic structure forming elements. With this practical production nanocomposite materials based on polymeric matrices may use available intermediates, for example, silicon and carbon-containing minerals, with the widespread use of high-dispersion technologies.

Based on the classical concepts of the mechanisms of formation of boundary layers with optimal structure, defining the parameters of strength, tribological, thermal, and others service performance composites and products from them, found that the main factors influencing the mechanism and kinetics of their formation, are the mechanical and energy. Mechanical factors contribute to the formation of the adhesive bond and the polymer matrix modifier by crushing fragments melt penetrated under pressure form the workpiece or products nanofields topography of the surface layer of the particle. Power factor provides the course of adsorption and orientation process at the interface "matrix - filler", leading to the formation of an ordered (quasi-crystalline) structure of the boundary layer with high strength characteristics of the parameters.

The combined effect of both factors makes possible the realization of synergies, providing a comprehensive impact modifier dispersed nanocomposite structure at the molecular, supramolecular and interfacial levels, which leads to a simultaneous increase in its parameters of strength, tribological, protective, etc. Service characteristics.

The physical criteria for assessing nanostate dispersed particles of condensed matter of varying composition and crystal structure. Studies Ajayan P.M., N. Kobayashi, Suzdaleva I.P., Trefilov V.I., Gusev A.I., Poole Ch., Owens F., Stroscio M.A., Dutta M. and employees, according to the determining role of size factor in moving the particles of condensed matter from macro to nano-state that they exhibit specific energy parameters influencing the processes of structure-modified matrix [9, 10].

Currently, there are no uniform sound approaches to the definition of the boundaries of existence dimensional nanoparticles. Boundary size of nanoparticles used in 100 nm has no clear physical basis and is not correlated with the experimental results.

Nanoparticles have at least two characteristic features (taxa). Firstly, the nanoparticles should have a developed surface. Second, the performance parameters in the nanoparticle physical properties (S) depend on its linear dimensions (r), i.e. nanoparticles of various substances exists function S (r). To characterize the surface of the nanoparticle concept is crucial. Nanoparticles regarded as an independent object which can be isolated from a mixture with other objects or other mechanical means. Therefore, the nanoparticles can be regarded as a kind of nanophases with its specific features.

The term "surface" for macroscopic and low-dimensional particle has a different physical meaning. For macroscopic objects 'surface' is treated as an imaginary infinitely thin film on one side of which there are atoms of a specific substance, and on the other side they are missing. The surface at the equilibrium state of the object is treated as a statistical system, neglecting the thermal vibrations of the atoms. In the transition to the surface of the nano-sized objects to be regarded as a dynamic system. Since the period of atomic vibrations immeasurably less time any experiment, it is necessary to speak of the surface layer. If we consider that in addition to the surface atoms mutual atomic configuration changes just adjacent to the outer nuclear layer, this surface layer can be commensurate with the size of the particle itself.

The average (idealized) dependence of the parameters of the physical properties of the particle size (S(r)) is shown in Figure 1.

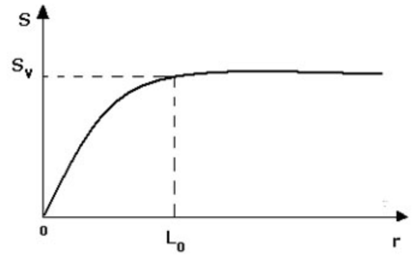

Figure 1 - The dependence of the parameter S of the physical properties of particle of size [S(r) function]

Figure 1 shows that a decrease in the size of the sample to a certain value $r = L_0$ the numerical value of the physical property $S(r) = S_V = const$, where S_V - value characteristic of the bulk sample (table). If the $r < L_0$ parameter $S(r)$ decreases monotonically and if $r \to 0$ then $S(r) \to 0$.

Analytical dependence of the function has the form

$$(1)\quad S(r) = \frac{S_V}{\left|\exp\left[\alpha\left(\frac{L_0 - r}{r}\right)^{3/2}\right]\right|} = \frac{S_V}{\left|\exp\left[\alpha\left(\frac{1-x}{x}\right)^{3/2}\right]\right|},$$

where L_0 - dimensional boundary between nanostate ($r < L_0$) and macrostate ($r > L_0$); α - parameter depending on the analyzed physical properties.

The presence of a sufficiently large volume of literature and our experimental data allows to confirm the consistency of the phenomenological formula (1). Obviously, if the parameter r passes through the dimensional limits L_0 of variation of the parameter S is not sharp. The more r different L_0, the more manifest the properties of the respective state. It can be concluded that when $r = L_0$ a substance is a change in the mechanism of physical processes or modified structural and energy characteristics of the particle compared to the bulk counterparts.

Based on the assumptions that the dimensional criteria nanostate L_0 should take into account the characteristic properties of substances that meet the basic understanding of the physical paradigm used tabulated values of the physical characteristics and is calculated by a simple formula, as the criterion for calculating the Debye temperature has been selected (θ_D).

Debye temperature (θ_D) as the L_0 parameter is not strictly a sharp boundary, but this option is entered in the modern physical paradigm. Debye theory of heat capacity indicates that the transition frequencies of quantum oscillators across the border ω_D there is a change in the mechanisms of the processes of substance. The atoms as quantum oscillators vibrate at a certain frequency. At low volumes on the geometrical dimensions of the frequency spectrum is influenced not only the structure of the substance, and the particle size. Vibrations of atoms to create a dynamic field that oscillates the electrons in the electron cloud with the same frequency.

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As part of the one-electron adiabatic approximation, we introduce the concept of Debye pseudo impulse P_D. We believe that the effective electron mass equal to the mass of a free electron. In this case

(2) $P_D = \sqrt{2mE_D}$

According to the formula of de Broglie Debye pseudo impulse P_D allows you to calculate the value of the Debye length (wave):

(3) $\lambda_D = \dfrac{h}{P_D}$

Consequently, the Debye length λ_D is the dimension of the boundary between the nano- and macrostate ($L_0 = \lambda_D$) substance in the condensed state.

Debye parameters ω_D, E_D, θ_D are related to each other by the equations [22]:

(4) $E_D = \hbar\omega_D = k\theta_D = \dfrac{P_D^2}{2m} = \dfrac{h^2}{2m\lambda^2}$

where $\hbar$, h ($h = 2\pi\hbar$) - Planck's constant, k - the Boltzmann constant, m - mass of the electron.

Note that in these terms the Debye length and the de Broglie wavelength coincide. Condition (4)

(5) $k\theta_D = \dfrac{\left(P_x^2 + P_y^2 + P_z^2\right)}{2m} = \dfrac{h^2}{2m\lambda_D^2}$

For an isotropic medium $P_x^2 = P_y^2 = P_z^2$. In this case, the parameter of the Debye length λ_D, for example, along the axis x (y or z) is defined by the expression:

(6) $L_0 = \lambda_D = \dfrac{\sqrt{1,5}h}{\sqrt{km}}\theta_D^{-1/2}$

After substituting the numerical values of physical quantities (h, k, m) received:

(7) $L_0 = 2,3\cdot10^{-7}\left[m\cdot K^{-1/2}\right]\cdot\theta_D^{-1/2}\left[K\right] = 230\cdot\theta_D^{-1/2}\left[nm\right]$

Analysis of the expression to evaluate the dimensional border nanostate particles of condensed matter using the Bloch theorem, the Schrödinger equation, concepts of Debye wavelength $L_0 = \lambda_D$ indicates eligibility to use it to determine the parameters of nanoscale particles (NSP) of different composition and structure.

The expression (7) suggests the existence of a probability of correlation between the critical size L_0 of the nanoparticles and the Debye temperature θ_D of the crystal from which they were formed.

The shape of nanoparticles can have a different habit (spherical, needle, plate). For such particles the parameter L_0 value will depend on the direction, i.e. there are cases where "nanofeatures" particles will not occur in all directions (x, y, z), and one (needle habit), two (lamellar habit) or three (spherical habitus). Therefore, by using modifiers particles having a relatively large size and nanometer active sites may be achieved by modifying effects inherent in the "classical" nanoparticles at least energy rich techniques for their preparation by synthesis or dispersing the intermediate product.

With the use of the proposed analytical expression (7) carried out calculation of critical parameter L_0 for particle single-element and multi-element materials - metals, non-metals, halides, semiconductor compounds. The results are in good correlation with literary sources, and allow us to determine the most effective technology for nanoscale modifiers for use in materials of polymer nanocomposites.

Analysis features nanostate particles of condensed matter using quantum theory allowed to show that for each substance there is an energy parameter E_0 differentiating the process. When $E > E_0$ dimensional effects do not play a role and true volumetric approach to describe the properties of the substance; when $E < E_0$ it need to take into account the size effects. The influence of the size factor on the energy characteristics of dispersed particles of silicon and carbon-containing components that have found the most widely used as a functional polymer modifiers and oligomeric matrix in the development of nanocomposites with enhanced service performance parameters.

The quantum-mechanical analysis of bulk and surface states of the particles showed that the potentials of the near-surface areas are not simply scrap the wave functions of the potential in the volume, but also have their own characteristics that appear in the manifestation of the electron work function of metals and a number of surface effects in dielectrics and semiconductors. The mechanism of formation of a particular energy state of the metal and dielectric samples, leading to the appearance of the surface mosaic observed experimentally.

The analysis of the crystal-formation mechanisms of nanoparticles in an active state by thermal or mechanical stress on the semi-finished products of natural layered silicates, frame and chain structure (mica, clay minerals, natural opals, tripoli). Among the most common are geosilikates laminates comprising various modifications of the mica and clay minerals. A common feature of this type is the presence of specific minerals of the crystal lattice with a perfect cleavage, consisting of layers of silicon-oxygen tetrahedra SiO_2, connected by a layer of metal-oxygen octahedra, and the interlayer cations.

With the destruction of the block layered minerals interlayer cations Na^+, K^+ to pass one of juvenile surfaces and form localized regions with a charge so-called " charge mosaic" such a structure of juvenile surfaces of layered silicates with charge areas, the area of which is many times the size of the cations causes the flow of intense processes of interaction with the environment of Nanophase particle. The presence of active centers on juvenile surfaces, form the basis of tetrahedra SiO_2, changes the course of adsorption processes on the surface of the particles and affects the structural ordering of molecules in contact with them and oligomeric polymeric macromolecules. Thus, the specific structure of the crystal-phyllosilicates creates the preconditions for the formation of nanoscale dispersion of the active particles, which have a modifying effect on the polymer matrix.

The actual particles of natural silicates are nonequilibrium structure, determine the existence of uncompensated charge. By EPR spectroscopy and TSC spectroscopy revealed the presence of uncompensated charge and the possibility of thermostimulated nanocuttent in powder samples of silicate minerals (opal, white and blue clay) (Figures 2, 3). Thermal effects on particulate phyllosilicates leads to a restructuring of their structure. On thermograms silicate minerals of various composition and structure can mention a few endoeffect at temperatures 373-423 K 523-623 K, 723-823K, 1023-1123 K, due to the processes of dehydration and dehydroxilation. The effect of 523-623 K characterizes the selection of interlayer water. When exposed to process blocks phyllosilicates mode thermal shock dehydration processes, water and interlayer separation degidroksilatsii occur virtually simultaneously, resulting in macrodestruction crystals due to rupture of the interatomic bonds. Product image type noncrystalline oxides and X-ray amorphous phases of different composition. The size of the particles formed during thermal processing products does not exceed 100 nm, according to modern concepts it allows to refer them to the nanoscale.

The special characteristic of the energy state and carbonaceous particulate matter (UDDG, carbon nanotubes, shungite). A characteristic feature of these low-dimensional particles of different composition, structure and technological history is the dependence of the energy state characterized by the magnitude of thermally stimulated currents, temperature. Therefore, when choosing the modifier to obtain nanocomposite materials with desired functional parameters (tribological, of strength, adhesion) must take into account the temperature range of manifestations of its maximum activity. Practical implementation of the established principles of the Energy of the respective components of nanocomposite materials based on polymeric matrices can achieve significant effects as in the doping content nanomodifier (0.001 - 1.0 wt.%), And the powers of filling 20 - 40 wt.%.

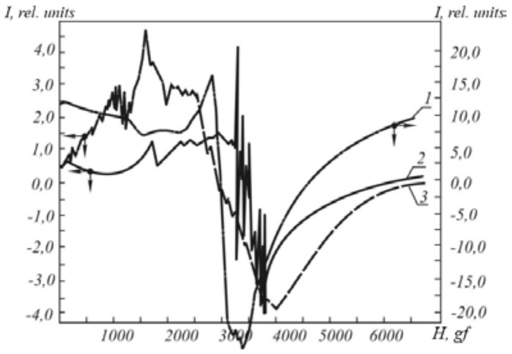


Figure 2 EPR spectra of opal (1) and clay minerals of different composition (2, 3)

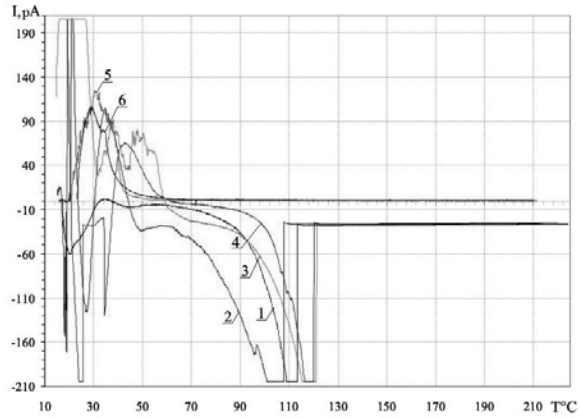


Figure 3 Characteristic form TSC-spectrum shungite source (1) and heat-treated at 100 ° C (2) 200 ° C (3) 300 ° C (4), 600 ° C (5) 800 ° C (6)

The mechanism of action of modifying nanoparticles in polymer matrices and different oligomeric structure. Based on the developed model, involves the formation of the adsorbed layer under the action of the active centers of the spherical nanoparticles form macromolecules, analytical expressions for calculating the concentration of modifier enough to transfer the entire matrix in an ordered state. The matrix will be in a modified form, if the nanoparticle influences the matrix macromolecule in the boundary layer thickness L:

(8) $$L = r_1\left[1+\frac{\rho_н}{\rho_n}\right]\left(\frac{1}{C_m}-1\right)^{1/3},$$

where r_1 - particle size.

Even if the content of doping nanofiller (0.001 - 1.0 wt.%) And the ratio of its modifying effect for at least 2 - 3 layers adjacent macromolecules achieved significant technical effect of improving the parameters of strength and tribological characteristics of nanocomposites based on thermoplastic matrices.

It is found that the efficiency of modifying action NSP affect not only the parameters of size, composition and structure, and form. When using a layered particles (flaky), whisker and the spherical shape of varying degrees of modification of the matrix M defined by the ratio of the total volume of the modified to the total volume of the composite. Taking into account the different energy states of various shapes NSP obtained degree of modification ratio of scaly (M_{sc}), whisker (M_w) and spherical (M_{sp}) particles at the same concentrations in the composite:

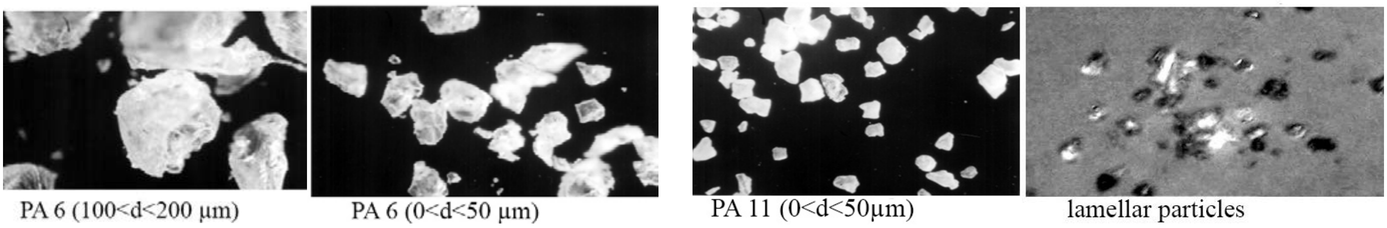
(9) $$M_{sc} : M_w : M_{sp} = 1:0,4:0,8$$

From this expression that when creating malonapolnennyh nanocomposites based on polymer matrices is the most effective use of layered modifiers, which include natural silicates - clay, mica.

The experimental and theoretical studies are based on the assumption of constancy of the structure, composition and habit NSP introduced into the polymer matrix. However, there are classes NSP (metal, oxide, metallic) that can be transformed under the influence of physical and chemical processes occurring in the boundary layers of the composites under the influence of operational factors (temperature, mechanical, mechano-chemical, etc.) To give the product a different composition and structure. Therefore informed choices NSP for modifying the target polymer or oligomer matrix involves a systematic analysis of the structural phase transitions with the energy, chemical and physical aspects of the formation and operation of metal-system.

A systematic approach to the analysis of the features of the modifying action of nanoscale objects of different composition, structure and technology for the development of scientific principles has led to the creation of functional engineering of nanocomposite materials based on commercially available thermoplastics (polyolefins, polyamides, polyacetals, fluoropolymers, and mixtures thereof) and technology of their processing in products with higher performance parameters.

To apply tribological and protective coatings of the developed nanocomposite materials based on polyamides developed the technology and installation for cryogenic grinding granular semifinished providing manufacturing powders with a given degree of dispersion of energy and activity (Figure 4).



PA 6 (100<d<200 μm) PA 6 (0<d<50 μm) PA 11 (0<d<50μm) lamellar particles

Figure 4 - The characteristic morphology of the particulate fraction of nanocomposites based on PA 6 and PA 11

The resulting powder semifinished provides functional coatings with the necessary operational parameters as they are formed by methods of the fluidized bed and flame spraying.

The compositions of tribological coatings used in the construction of automobile units to improve the wear resistance of splines driveshafts transport and special equipment, the protection of the elastic members of brake chambers trucks from corrosion and mechanical wear and fatigue failure and to prevent jamming in the mechanism of the drive lathe chuck technology metalworking equipment. Application of nanocomposites based on polyamide 6 (JSC "GrodnoAzot") instead of imported counterpart PA 11 («Rilsan») provided to obtain significant economic effect on the JSC "Belcard", JSC "BelTAPAZ 'products are used for the assembly of automotive engineering and metalworking equipment produced in the Russian Federation [11].

For the manufacture of nanocomposite materials based on polytetrafluoroethylene developed the original tooling and equipment for the implementation of effective activation of components in the process of filling the system of training, cold pressing, hot monolitization and calibration. The technological methods to eliminate or minimize the negative effect of structural factors at various levels, enhancing defect nanocomposites and products of them as a result of the clustering of the particles of the reinforcing filler (HC) and their inefficient wetting polymer binder being in viscoplastic state at temperatures monolitization (573 - 673 K) pieces .

The effectiveness of the developed nanocomposite materials based on polytetrafluoroethylene due to a decrease in the content in the composite costly filler - carbon fiber (CF), while maintaining the required performance parameters and a decrease in energy consumption due to optimization of the technological cycle of manufacturing products (billets) . In addition, the use of technology Cold monolitization to reduce the loss of the composite due to allowances. Application of the developed nanocomposite materials for the manufacture of elements moving seals compressor equipment for compressed and liquefied gases (JSC "Grodno Azot", JSC "Sumy Scientific-Technical Center", JSC "Frunze Sumy Machine Building Scientific Production Association") provides an increase in the service life of less than 2 times [14-16].

Increased resistance to spike universal joint propeller shaft brinnelling achieved by using plastic lubricants designed for heavy duty units containing polymer fibers and low-dimensional nanoporous carbon particles or metal particles thermoplastics. Developed based lubricant base oils and industrial-20, I-40 at ANTI characteristics and resistance to mechanical degradation far superior lubricant used CIATIM 201, №158, Lithol-24 [16].

To ensure the technical performance of the brush attachment drive road cars developed abrasion composite material based on PP-doped doping additives (0,01-2,0 wt.%) Geomodifiers layered, chain or frame structure. Introduction into the polymer matrix of the active particles geosilikate reduces melt index composite 1.5-2.0 times and stabilizes the extrusion of strands at the multislot head. The structure of the composite with physical crosslinks oriented uniaxial stretching with the formation of the ordered area, increases the strength of 1.3-1.5 times and 1.5-2 times material to the resistance to abrasion. Designed nanocomposite composition allowed to apply to PP as the base material for general technical purposes instead copolymer doped with a thermoplastic elastomer having almost twice higher value [16].

Conducted testing of the developed nanocomposite formulations of engineering materials based on thermoplastic matrices at industrial enterprises of Belarus and Russia testifies to their high efficiency and expediency of application in the construction of automotive engineering, process equipment and valves.

4. Conclusion

Based on a systematic approach to the study of the impact of the structural features and the energy state of nano-sized particles of condensed matter the mechanisms of physical and chemical processes in polymer matrices that define the parameters of their structure at the molecular, supramolecular and interfacial levels. Methodological approaches to the creation of nanocomposite materials with high engineering parameters of strength, tribological, adhesive and protective characteristics on the basis of industrial thermoplastics and their production technology and processing into products.

Methodological principles of obtaining nanocomposite materials based on thermoplastics industry, consisting in:

- crystal-established assumptions selection of natural and synthetic carbon-containing, silicon metal and semi-finished products for directional formation of active nanoparticles with desired structural and morphological and energetic parameters for optimum technological impact (mechanochemical, thermal, laser);
- implementation of the conditions prevailing energy nanomodifiers compliance mechanism of the optimal structure of the polymer, oligomeric matrices and combined at different levels - molecular, supramolecular and interfacial;
- ensuring the development conditions of the preferred mechanisms of interfacial physico-chemical interactions of the components with the formation of boundary layers optimum structure, defining mechanisms of destruction of the nanocomposites under the influence of various operational factors [11-16].

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