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Current Status and Prospects for the Use of Nanosystems as Protective Coatings: A Review

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This article is concerning to a review of experimental studies of nanocomposite-coatings' formation processes with improved functional properties, as well as the study of nanocrystals' formation patterns within the bulk of binary and ternary alloys. Among the most popular materials of the present time, the main place is occupied by composite coatings, to improve functional properties of which, the structuring of the metal matrix with nanodispersed particles is used, as well as binary and ternary alloys with a nanocrystalline structure. Such coatings have found application in microelectronics, mechanical engineering, aerospace and rocketry, chemical industry and environmental technologies. Introduction of nanosize components into the coating composition makes it possible to obtain catalytic, anticorrosion, magnetic, and high-strength materials, which are in demand in aggressive environments and high temperatures.

Дана стаття стосується огляду експериментальних досліджень процесів формування нанокомпозитних покриттів з поліпшеними функціональними властивостями, а також дослідження закономірностей утворення нанокристалів в об'ємі бінарних і потрійних стопів. Серед найпопулярніших матеріялів сучасности основне місце посідають композиційні покриття, для поліпшення функціональних властивостей яких використовується структурування металевої матриці нанодисперсними частинками, а також бінарні та потрійні стопи з нанокристалічною структурою. Такі покриття знайшли застосування в мікроелектроніці, машинобудуванні, аерокосмічній і ракетно-космічній техніці, хемічній промисловості й екологічних технологіях. Введення до складу покриття нанорозмірних компонентів уможливлює одержати каталітичні, антикорозійні, магнетні та високоміцні матеріяли, затребувані для використання в аґресивних середовищах і за високих температур.

Key words: coatings, nanocomposites, structure, applying.

Ключові слова: покриття, нанокомпозити, структура, застосування.

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1. INTRODUCTION

One of the most important purposes of modern materials science is the study of patterns formation nanocomposite materials microstructures [1–9]. This is due to special attention to fundamental problems of composite systems creating and application in various fields of engineering and technology. Therefore, the development of fundamentally new physical and technological stages of multicomponent systems structures formation is one of main problems in the field of nanomaterials and nanotechnology science [10–13].

It is well known that composite materials are heterophase structures, taken from two or more elements, preserving the properties of each component. Most solid-state composites are a matrix, in the volume of which the second phase is distributed in a certain way. Matrix elements can be metals and their alloys, inorganic and organic polymers, ceramics and other materials [14, 15]. Strengthening or reinforcing components are both thread-like fibres and fine particles of various natures.

In recent years, scientists from leading countries of the world determined that nanostructured materials such like nanotubes, fullerenes and nanoparticles are the most promising in terms of improving functional properties materials by several orders of magnitude [16].

Among the most popular materials of the present time, the main place is taken by composite coatings, to improve functional properties of which, structuring is used with metal-matrices' nanodispersed particles, as well as double and triple alloys with nanocrystalline structure. Such coatings have found application in microelectronics, mechanical engineering, aerospace and rocketry, chemical industry and environmental protection technologies. Introduction to composition coatings nanosize components allows obtaining of catalytic, anti-corrosion, magnetic and high-strength materials, which are in demand in aggressive environments and high temperatures.

Thus, the study of nanosystems and their influence on the physicochemical properties patterns formation is one of the pressing problems in the production of nanostructured composite coatings.

Today, one of the promising areas for improving the performance characteristics of industrial products is modernization-using coatings based on transition metal nitrides and intermetallic compounds. An option was proposed for converting such protective coatings into a nanostructured state, as well as the formation of multilayer structures with nanoscale architecture [17–20]. The gradient nature of multilayer structures allows for a variety of functions, such as increasing adhesion, increasing resistance to wear and corrosion, and much more. Alternating two or more layers of materials with different physical and mechanical characteristics makes it possible to change the properties of the system, including stress concentrators, and influence the propagation of cracks as a result of achieving increased fracture toughness of the material [21–23]. Multilayer compositions based on binary nitrides TiN/CrN, TiN/MoN, ZrN/CrN, ZrN/MoN, ZrN are of particular interest.

The literature typically uses hypotheses and ideas from various fields of natural sciences, namely, solid-state physics, physical materials science, chemical physics and condensed matter physics. Multilayer nanocomposite coatings are of particular interest because they have some special physical, chemical and micromechanical properties, such as high temperature stability [23], hardness and improved thermal barrier properties, which are most suitable for operation under extreme tribological conditions [24–28].

At the same time, the properties of nanocomposite materials are not always necessary in the entire volume of structure or part, but only in a thin layer of a certain thickness. In this aspect, the method of composite electrolytic coatings (CECs), which is based on the electrolytic precipitation of metals and dispersed particles, makes it possible to obtain compositions in the form of thin protective layers is promising.

The CECs' method, compared to others, is much less labour-intensive, does not require high temperatures and is economical. These advantages of the CECs' method led to the intensive development of research work in the U.S.A., Germany, the CIS, and other countries.

The growing interest in nanocomposite electrolytic coatings is evidenced by publication activity over the past 5 years and citations in both applied and basic science.

2. METHODS OF OBTAINING CEC

Methods for obtaining nano-CECs can be divided into two main ones groups. The first of them includes methods in which standard electrolytes add nanosize powders and as a result get a metal coating in which nanoparticles are embedded in the matrix. The second group includes deposition methods, in which, during the formation, electrolytic coatings use various electrolysis modes, and thus, purposefully reducing the crystal size down to obtaining an amorphous-crystalline structure.

During the process of electrolytic coatings, as a second-phase forming powders with particle sizes not more than 100 nm were used. These CECs are accepted count nanostructured coatings, and technology for their fabrication, namely, nanotechnology. For example, in work [29], a multichamber electrolyser is used for application of nanocomposite Cr_3C_2–TaC–NiCr coatings on steel substrate A29 1044 (Fe–0.45C–0.18Si–0.6Mn). According to literary data, it was found that developed method provides conditions to form dense layer nanocompositions with porosity below 2% and microhardness 800 ± 25 $HV_{0,3}$.

The authors' works were carried out [30, 31], in which nanocomposite Ni–Al_2O_3 coatings were prepared method jet electrodeposition and analysed influence of Al_2O_3 concentration in the electrolyte current density and speed of electrolyte stream on the content of nano-Al_2O_3. In work [32], microstructures of composite coatings were studied using methods electron microscopy. The authors assessed the effect of Al_2O_3 nanocontent on microhardness and corrosion durability. According to the results, it was established that deposited Ni layers have nanocrystalline microstructure with average 50 nm in size. Al_2O_3 particles are well dispersed in the layer, and the content particles is up to 12.2 at.%, and also presented composite Ni–Al_2O_3 coatings, which were obtained in a new way from modified Watt type electrolyte containing Al_2O_3 nanoparticles, where instead of mechanical stirring a strong magnetic field was created in a direction parallel to the electrolytic current. The authors investigated influence of magnetic field on particle content, morphology surfaces, microhardness and wear resistance. It was discovered that strong magnetic field plays an important role in the formation of composite coatings. The number of Al_2O_3 nanoparticles in the composite coating is increased with increasing magnetic-flux density, reached its maximum values at 8 T and then decreased slightly.

Microhardness and wear resistance nanocomposite coatings also increases with increasing magnetic flux density compared to purity of Ni coating. This is because co-sedimented Al_2O_3 nanoparticles were evenly distributed in the Ni matrix and contributed significant increase of microhardness and wear resistance of composite coatings.

Nanocomposite coating Ni–Al_2O_3 obtained by electrochemical precipitation has many excellent properties and is a promising material for use as a protective coating. In this study, Ni–Al_2O_3 nanocomposite microcomponents were prepared using different electric current densities [33].

Young's module was calculated from experimental curves bending composite cantilevers. Research has shown that Young's module of Ni–Al_2O_3 nanocomposite amounts to approximately 202 GPa at 45°C its deposition and current density of 10 mA/cm^2, but at current density of 60 mA/cm^2, sharply drops to 67 GPa. By compared to electrodeposited pure Ni, the Ni–Al_2O_3 nanocomposite exhibits increased elastic properties [34].

Ni–Al_2O_3 nanocomposite coating was prepared by electrolytic rubbing, and for comparison, Ni–Al_2O_3 coating was also applied by electrolysis. It was studied the morphology of surfaces, microstructure, microhardness and wear resistance coatings [35].

The results showed that Ni–Al_2O_3 nanocomposite coating, obtained by electrolytic rubbing has a smoother, fine, dense microstructure and more uniform characteristics across compared with the electrolytic nanocomposite coating Ni–Al_2O_3.

The first demonstrated more high microhardness and better wear resistance under lubrication conditions compared to the latter at tribological tests [36].

Joint deposition Cu (Zn) and graphite powders size less than 10 nm [37]. Electrocomposite coatings were cooked with a help of electrodeposition traditional methods. The influence has been studied parameters such as stirring speed and current density, on compound electrodeposited brass, as well as the influence of graphite particles concentration in brass bath, air-mixing speed to include graphite in brass matrix and deposition efficiency.

Wear resistance and friction properties of compositional Cu (Zn)–C coatings containing different percentages of graphite particles have been studied. The presence of graphite particles improved antifriction properties composite coatings. It has been shown that composite coating, containing approximately 3.7 vol.% graphite, has the best tribological properties.

Nanocomposite Ni–SiC coatings were received electrodeposition from nickel sulphate bath containing nanoparticles SiC with average size particles 30 nm. The characteristics of the coatings were assessed using scanning electron microscopy and test on microhardness.

Friction and wear characteristics nanocomposite Ni–SiC coatings and Ni films have been comparatively studied during the process of sliding on ceramic Si_3N_4 balls under non-lubricated conditions. The results showed that according to compared to Ni nanocomposite film Ni–SiC coating shows increased microhardness and wear resistance [38–41].

3. PROPERTIES OF MULTILAYER COATINGS

An important role in the formation of nanostructured films and coatings plays a role in controlling the crystallographic orientation and grain sizes by changing the energy of particles during deposition and mass transfer, introducing individual elements into composite solid matrix or variations of nanometer-thick layers with alternating amorphous and crystalline phases. In this case, microstructure of film is significantly affected by the temperature of the substrate and the energy of ions, which transfer their kinetic energy to the coating during deposition [42]. This specificity is characteristic primarily of the magnetron sputtering method, in which the energy of the deposited particles depends on working gas pressure in the vacuum chamber, the distance from the source to the substrate, and magnitude of the bias potential applied to the substrate.

Vacuum-arc deposition is highly ionized technology using the ion energy of about 10 eV, which makes it possible to regulate zonal temperature and kinetics of interaction with the cathode spot, from which the emission of ions and neutral particles involved in the formation of coating takes place. Consequently, the structure and substructure significantly depend on the particle collision energy, and by changing a set of parameters, it is possible to establish the preferred range of physical and mechanical properties of coatings [43]. The main criteria for nanocrystalline films desired properties obtaining grain size and crystallographic orientation, which is provided by ion bombardment during deposition. The effectiveness of ion bombardment is manifested in a decrease in crystallite sizes, relaxation of compressive stresses, compaction of grain boundaries and localization of radiation defects, which leads to changes properties coatings [44, 45]. At this optimal method for control of crystallite sizes in the direction of coating growth is the formation of a multilayer architecture with nanoscale layers. This coating structure is ensured by periodic deposition of thin layers of various compounds of a given thickness.

The work [46] presents classification of hard coatings according to the type of their connections (Fig. 1). Nitrides, carbides and carbonitrides transitional metals (especially elements from groups IV*B*, V*B* and VI*B* of the periodic system) are among the most favourable materials for creating wear-resistant coatings.

The structural and compositional distribution in a multilayer coating relative to individual layers, interphase boundaries, as well as their dimensional relationships allows setting the functionality of modernized surface, controlling its properties, such as hardness, strength, elasticity, *etc*.

During the deposition of a multilayer structure, the proportion of interphase boundaries increases relative to the total volume of the interfaces, which prevents propagation of dislocations and cracks, thereby raising mechanical resistance systems. However, the reduction in the thickness of resulting layers is limited to values, upon reaching which blurring of the interphase boundaries occurs [47,

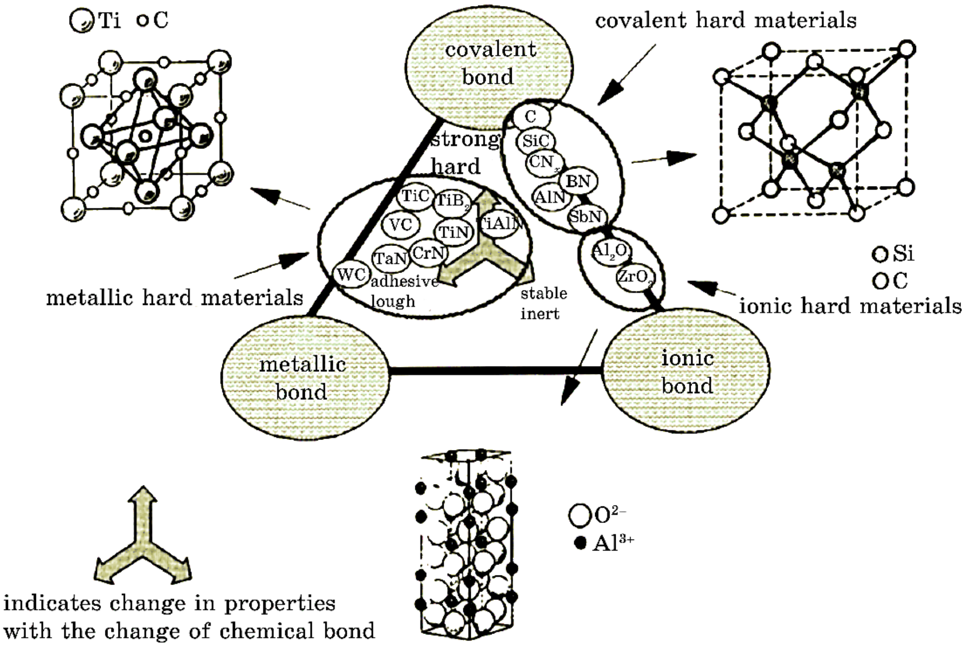

Fig. 1. Classification of different groups of hard coatings according to their bonding type and crystal structure [46].

48]. Therefore, studies of the phase state, structure and physical and mechanical properties data nanosize complexes present special scientific and practical interest.

According to the reverse Hall–Petch effect [49], in nanocrystalline materials with grain sizes of about 10 nm, maximum hardness values are achieved. Grain strengthening is happening behind due to intercrystalline processes, such as absorption by the grain boundary of the dislocation core and interaction with point defects [21, 50].

In works [51, 52], the influence on the mechanisms hardening size nanograins and near-surface effects at the film–substrate boundaries was investigated. There is also a strong dependence of the structure and properties of coating on technological parameters of its production, which complicates the unambiguous interpretation of the nature of grown structures. At interphase boundaries, atoms of various thin layers crystal lattices in multilayer films can be replaced, which in turn leads to the appearance of additional deformation energy proportional to the shear modulus of material. Layers with different shear module prevent the movement of dislocations. In addition, trajectory deviations or redistribution of dislocations and cracks at grain boundaries help to increase the resistance of coatings to fracture. Periodic alternation of nanometer-scale layers with different physical and mechanical characteristics makes it possible to change significantly such properties of a multilayer architecture as the concentration of internal stresses, crack propagation and, consequently, an increase in viscosity destruction (crack resistance) of the structure [44].

Particular attention should be paid to the issue of existing mechanisms and their modified versions. During research, a series of experimental works was carried out to study the mechanical, elastic, and plastic properties of multilayer systems using the example of formation and propagation of cracks and energy dissipation [46, 52].

Mechanical loads produce multiple effects on the multilayer structure, which are divided into surface effects (cracking), deformation of the internal layers of interphase boundaries, as well as changes at the grain boundaries and response of the entire coating. A crack begins on the surface, can branch and change its trajectory at grain boundaries (strengthening due to grain boundaries), similar processes also occur at the interfaces between layers (strengthening due to interphase boundaries). On the other hand, local delamination may occur at the interfaces due to occurrence nanosize cavities, which lead to local relaxation of internal stresses and an increase plasticity on nanolevel. In addition, there is an interaction between cracks and periodic deformations at the interfacial nanoscale boundaries multilayer structures, as well as the interaction of through cracks with the substrate material. These mechanisms are shown schematically in Fig. 2 [46].

If we look at this process from the dislocation motion point of view, then, due to the presence of a large number of grain boundaries in the nanocomposite coating, dislocations, which propagate through the grains, begin to slow down their movement at the grain boundaries, which leads to their accumulation. Consequently, for their further movement, increased voltage is required, which leads to an increase in hardness [53]. The dislocation density increases with decreasing grain size, which means the hardness increases proportionally size nanograins. However, it is extremely important to note that with very small grain sizes (less than 2–3 nm) the universal mechanism accumulation of dislocations is unacceptable because dislocations cannot be generated in small grains. It is believed that grain-boundary sliding and grain-boundary migration play an important role in the reduction of hardness at small grain sizes, better known as the Hall–Petch inverse effect [49].

Internal interphase boundaries demonstrate a positive effect on the micro- and macroscopic properties of the entire coating, when their number is carefully selected relative to the total volume of coating.

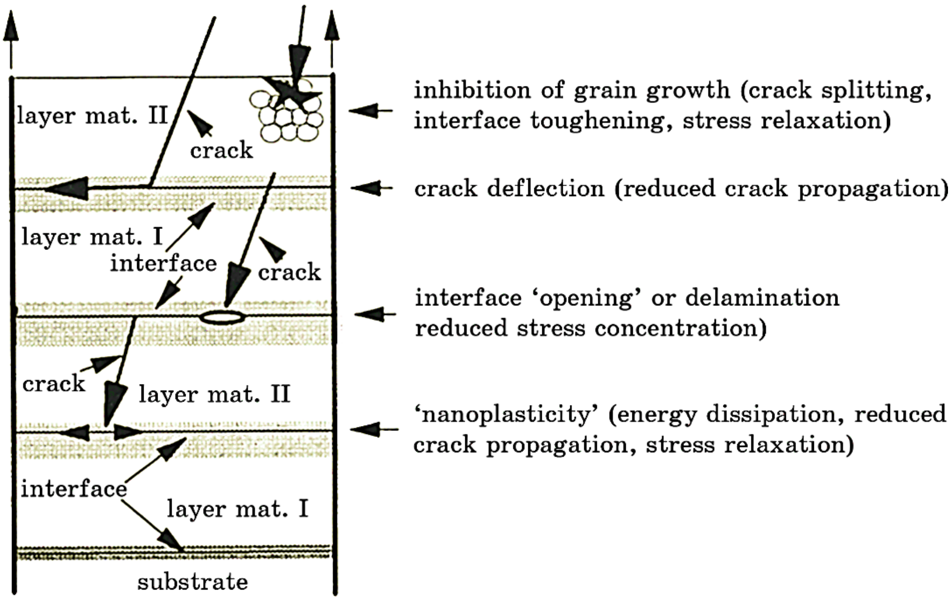

Fig. 2. Conditional mechanism of crack propagation in multilayer systems and their distribution at interfacial boundaries [46].

In addition to grain geometry, significant influence on physical and mechanical properties is determined by their crystallographic orientation. In the case of applying single-layer coatings, orientation of grains is largely determined by the material and orientation of substrate [18, 54]. In nanocomposite structures, there is an increased interfacial region, which makes the contribution of surface and interfacial energy dominant. This improves hardness, strength, and wear-resistance compared to single-layer counterparts [55]. By manipulating the ratio of layer thicknesses and grain size, tendency to change the surface energy can appear, therefore, transition to one or another preferred orientation is possible. In this case, the first (template) layer will determine orientation subsequent recrystallizing during the process layer growth. For example, in work [56], dedicated to NbN/MoN nanocomposites, crystallographic orientation switched from (111) to (200), which has a lower surface energy, as soon as the thickness of molybdenum template layer exceeded 2.1 nm. In Ref. [57], it was revealed that in coatings TiN/Si_3N_4 at decrease grain size from 50 to 20 nm or less, which was achieved by increasing the silicon concentration, the structure changed from (111) to randomly oriented, which made it possible to regulate the wear resistance and hardness of resulting coatings.

As a result, several approaches can be proposed to explain mechanisms effect superhardness, including blocking dislocations at interfaces, differences in the nature of internal deformations in individual layers and an increase in the reverse Hall–Petch effect. Moreover, all the considered models have common premises to achieve superhardness. First, two composite layers must have different shear moduli and, therefore, different dislocation line energies. Secondly, layers must be thin enough to limit the formation and movement of dislocations in individual layers. Finally, and most importantly, two modulation levels must form consistent interfaces [58–62]. In the case of a multilayer coating that meets these criteria, to increase hardness, significant internal stress is required to move the dislocation along the interface.

4. MECHANISMS OF COATING FORMATION

Analysis of literature data allowed classifying the nanocoatings developed to date as follow [63–67].

1. Coatings with nanosize grains and crystals, unlike classic ones, have increased microhardness and plasticity. The behaviour of nanocrystalline materials with grain sizes of 10 nm or less is determined mainly by processes in the boundary regions, since the number of atoms in grains is comparable or less than in their boundaries. This circumstance significantly changes the nature of the interaction between neighbouring grains, for example, it inhibits the generation of dislocations and prevents propagation of cracks due to the strengthening of grain boundaries. Under these conditions, there are no dislocations in nanograins.
2. Nano-CECs are characterized as a solid nanocrystalline phase, which consists of grains with a thin layer of an amorphous phase. Consequently, solid grains of strengthening phase are separated from each other by thin layers of another-phase atoms. Such coatings have ultra-high hardness, high elastic return coefficients, and high heat resistance.
3. Nanolayer coatings are highly crack resistant. Conditions for their production appeared in recent decades, when multilayer coatings containing titanium nitride/niobium nitride, titanium nitride/vanadium nitride, aluminium nitride/chromium nitride with alternating metal and composite nanolayers were synthesized. Such coatings differ in elastic modulus and thermal expansion coefficients. The thickness of layers should be so small that new dislocations cannot arise within one layer. At the same time, linear defects accumulate at the boundaries of soft layers of coatings and form elastic stresses in harder layers. Total number of layers in nanocoatings is calculated, depending on the technological tasks, and, in some cases, can reach up to two hundreds.

In work [71], Ukrainian specialists of the Kharkiv National University showed an analysis of the current state of research into the structure and properties of solid (hardness $H_\mu = 40$ GPa) and superhard ($H_\mu > 40$ GPa) nano-CECs. The review examines physical principles of thin coatings hardness increasing (< 10 nm), methods for their creation, possible uses and the scope of further development of this nanotechnology area. The authors emphasize that information about the hardness of certain nano-CECs exceeding the hardness of diamond and boron nitride needs to be confirmed, as they may be the result of methodological errors, and emphasize the need to develop new non-destructive hardness measurement techniques. They also present a set of experiments to explain the physical factors involved in increasing the hardness of nanocoatings.

Micrograined coating structure based on the principles of compounds of refractory metals is explained by high hardness and wear resistance. Nowadays, methods are emerging for producing modern vacuum nanocoatings with grain sizes of no more than 100 nm; this type of coating has increased mechanical characteristics [72].

Over the past few years, the influence of nanostructure on the properties materials was often discussed and some synthesis results were shown as volumetric reinforced materials nanostructured elements, and surface layers. Nanostructured pure and alloyed superficial Ti–Ni layers for medical tools were developed. It is common knowledge that alloying Ti–Ni with various components allows controlling temperature and martensitic transformation for subsequent use [73–77]. Due to the content alloying element, phase transformation sequence and possibility of repeated temperature cycles ternary Ti–Ni–Cu alloys occupy special place among all multicomponent alloys. Results studying impact nanodispersed phases on features of compositional materials with shape-memory effect (SME) make it possible to obtain fundamentally new results to improve performance properties superficial layers nanostructured memory effect materials forms.

Nanodispersed powders are widely used for structuring of metal matrix apply that obtained by mechanical and mechanochemical activation methods. Authors have chosen two-component Ti–Ni and Ni–Al powders' plasma method as a way of surface layers materials functional characteristics increasing, because it allows the formation of amorphous–crystalline layers, has high performance and versatility.

Experimental setup for plasma electrolytic deposition (PED) made it possible to control all stages of surface modification, including temperature and mechanical processing. Based on analysis and generalization of experimental results optimized design of the VPS-3*D* installation was developed, allowing to obtain specified technical and structural characteristics of coatings.

It is noteworthy that in this method of nanocomposite coatings deposition defining properties are both dimensions and nanodispersed powders energy intensity.

Because of crushing, local tensions, which in the process of further relaxation ultimately change the properties of the dispersed phase as a whole. According to modern ideas, the main reason for modifying powder characteristics is to increase temperature and dislocation energy, as well as an increase in interphase boundaries.

Original size of dispersed TiNi particles was of 2.0–12.0 microns. Because of mechanical activation during four hours, average size of nickelide-titanium powder particles decreased up to 50–100 nm.

Based on the analysis of experimental data, it was found that the average size of ПН55Т45 powder components after 2.5 hours of grinding is 0.9–7.0 nm, 93% of which are up to 5.0 nm in size in the form of flat disks with a thickness-to-diameter ratio of 1:7. Thus, more than 90% of the ground powder is nanosize dust. Analysis of the morphology of the composite coating obtained by plasma spraying of microsize and nanosize Ti–Ni and Ti–Ni–Cu powders made it possible to establish that the use of mechanically activated dispersed phase leads to a significant reduction in porosity and improvement in adhesion by 7.51–10.78 times. Obviously, this is due to the accumulation of energy by the particles, which contributes to their better penetration into the surface layers of the material, which ultimately leads to the formation of a monolithic, almost pore-free microstructure with improved mechanical properties [74].

During the grinding process, particles undergo intense plastic deformation, which ultimately leads to the formation of defects, which in turn become centres for the formation of nanocrystals. Chamber grinding attritors are accompanied by increase temperature. Mechanical activator camera cools down flow-through water to room temperature. Rapid cooling in the area of contact of powders with the working fluid contributes to increased fragility and formation nanocrystals. This process of nanoparticles' formation occurs until nanograins' dimensions achieved critical sizes, at which further plastic deformation will not affect the process. Analysis of experimental-studies' results led to the conclusion that as a result of the formation of numerous defects, accumulation occurs energy inside nanopowder particles, which stands out in the process plasma spraying and promotes the best adhesion of nanostructured compositional coatings [75–80].

5. CONCLUSION

In the presented review, class of nanosize coatings with nanolayer architecture, consisting of both binary and multielement (of 3–6 elements) nitrides, carbides and their combinations, was considered. It was shown, that extremely fine-scale structural ordering on nanolevel is prerequisite for the design of new multifunctional hard surfaces. The review does not pretend to be a complete analysis of numerous works (the number of which reaches several thousand); however, we believe that the main patterns that appear in these structures (systems) have been considered, and we can briefly outline them. Regarding further progress in the development of multilayer coatings with alternating nanolayers of various functional purposes with understanding the role of the selected materials, growth conditions, microstructure and required properties, a systematic approach is required. Changes in the columnar structure and grain shape, as well as coherence between layers, can lead to significant changes in physical and mechanical properties.

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