Research Article ISSN 2832-9384

Journal of Materials and Polymer Science

Tribological Characteristics of Nanocomposite Coatings Based On the Principle of Multielementality

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Submitted: 24 Feb 2026; Accepted: 10 Mar 2026; Published: 17 Mar 2026

Citation: Janahmadov, A. Kh. & Javadov, M. Ya. Tribological Characteristics of Nanocomposite Coatings Based On the Principle of Multielementality. *J mate poly sci, 6*(1) :1-6. DOI : https://doi.org/10.47485/2832-9384.1082

Abstract
The paper examines the ability to control a number of tribological and mechanical characteristics of coatings that provide optimal ratios of hardness to effective modulus of elasticity based on the principle of multi-element. The indicated values of ratio provide a high value of hardness and low values of the coefficient of friction and wear. In addition, reduction in the difference of the elastic modulus of the substrate and the coating allows to increase the adhesive properties. Criteria for transient contact conditions are given, as well as the values of the ratio of hardness to elastic modulus, which characterize the resistance of the coating to plastic deformation. The results of the study can be used in friction units, especially in aerospace machines, mechanisms and instruments.

Keywords: tribology, nanostructured coating, friction coefficient, wear coefficient, criterion of transition of the contact mode, hardness, effective modulus of elasticity.

Introduction

The development of science and technology, technology has become more sophisticated and new classes of technologies have appeared, such as nanotechnology, for which the problems of experimental research, modeling, calculation and optimization of technological processes, ensuring their efficiency and product quality, are becoming increasingly relevant. Particularly important is the solution of problems for modern high-tech technologies arising under the action of mechanical loads and/or physical fields and chemical active media that allow the design and manufacture of both materials and directly products and structural elements with a given multi-level structure and the required distribution of mechanical and physical chemical properties.

A wide variety of problems in the mechanics of a deformable solid and associated physical and chemical problems are associated with the technologies of nano- electronics and micro-electronics. The development of mechano-physico-chemical models and methods for calculating a number of technologies of nano- electronics and micro-electronics contributed to their development and optimization (Валиев et al., 2005; Гольдштейн & Морозов, 2012).

In connection with the priority development of nanotechnology, the question arose of applying methods of continuum mechanics to study the mechanical properties and phenomena mainly of deformation and fracture in nanostructures. These structures are characterized by two features that may impede their study. Firstly, it is the discreteness of the medium at the nanoscale, which contradicts the basic position of mechanics on the existence of an elementary infinitesimal volume dV, which, on the other hand, contains a very large number of atoms and molecules. This allows us to introduce the concept of mass density, energy, etc. Secondly, nanostructures are characterized by the fact that the dimensions in one of the directions in space are from 10 to 100 Nm, and the task of nanotechnology is to create objects up to 1000 Nm in size (Роко et al., 2002). Therefore, these structures have an extremely developed surface, the dimensions of which cannot be neglected in comparison with the dimensions of the "bulk" part. It should be borne in mind that both subsystems have very different properties. Thus, it is necessary to directly compare the results obtained in the framework of continuum mechanics and the molecular dynamics method, which should be considered as a test case.

Two approaches are feasible when applying methods of mechanics of a deformable solid to nano objects (Роко et al., 2002). The first one is the consideration of a nano-object as a single "elementary" volume with properties averaged over the subsystems "volume" - "surface". In this case, further fragmentation of the entire system into elementary volumes (or the construction of a numerical grid inside a nanostructure) for carrying out numerical calculations is illegal. The second approach is to consider the nanostructure as a heterostructure. In this case, using the characteristics of the "surface" and the interface obtained within the framework of the method of the molecular dynamics, it is possible to calculate the process inside the nano object.

The scale effects are critical in the mechanics of nanostructures, in contrast to microstructures. Since the mechanical behavior of structures at micro levels differs from that observed at nanoscale levels, the modified theories of continuum mechanics applied to microstructures differ from the corresponding theories in nanostructures (Lei et al., 2016). Generally, the hard stiffness is observed at microscale levels, while the soft stiffness usually occurs in the mechanics of nanostructures. Therefore, the size-dependent models, including mutual stress (Ghayesh & Farokhi, 2017; Farokhi & Ghayesh, 2018) and elastic stress gradient (Akgoz & Civalex, 2013), are often used in the analysis of the mechanical behavior of microstructures, including micro beams, micro bars and micro plates, while the nonlocal theory of elasticity Farajpour et al. (2018) is used in nanoscale structures. A more generalized size-dependent model based on continuum mechanics, which can be used to predict size effects at various small scales, was obtained in Lin et al. (2015) by combining these modified theories.

Due to the difficulties of performing accurate experimental measurements at the nanoscale level and the high computational costs associated with the modeling method of molecular dynamics, the modeling of nanostructures using continuum mechanics has recently attracted increasing interest (Farajpour et al., 2018).

Purpose of work: by controlling the tribological and mechanical characteristics of the coatings, to ensure the optimum ratio of hardness to the effective modulus of elasticity based on the principle of multi-element and identify criteria for transient contact conditions. For this, it is necessary to significantly reduce the friction coefficient, increase the adhesion and hardness of the coatings during the observed contact interaction of solids.

Problem formulation. Surface engineering is one of the most promising and rapidly developing areas of modern materials science, serving various fields of science and technology - physics, chemistry, medicine, mechanical engineering, metallurgy, etc. Currently, a new branch of tribology is developing - nanotribology, combining experimental and theoretical studies of friction, wear, lubrication, chemical activity and tribo-electromagnetism of the surface at the nanostructured level. Such an integrated approach is useful for solving an important task of modern materials science - the creation of nanostructured metal materials, the surface of which has both lubricating and anti-corrosion properties.

In recent decades, a research direction has been intensively developing related to the creation of nanocomposite coatings consisting of several at least two phases with a nanocrystalline and/or amorphous (a-) structure (Musil, 2012; Коротаев et al., 2011; Korotaev et al., 2013; Martinez-Martinez et al., 2009; Lüth, 2015; Frey & Khan, 2015; Janahmadov, 1996).

To date, nanocomposite coatings have been created (based on nitrides and carbo - nitrides of transition metals (Musil, 2012; Коротаев et al., 2011; Korotaev et al., 2013; Janahmadov & Javadov, 2016; Janahmadov, 2020), which have super-hardness (>40GPa) or high hardness (>25GPa). Coatings of this type are widely used in the manufacture of cutting, and processing tools, providing a significant increase in tool life (Musil, 2012). However, super-hard coatings are much less effective in protecting conventional parts operating in friction units of engines and mechanisms, due to the relatively high coefficient of friction, the tendency to peeling (low adhesive and cohesive strength), to brittle In this connection, nanocomposite coatings with a combination of mechanical and tribological properties (high hardness, wear resistance and low friction coefficient) are of interest for a significant increase in the service life of parts operating in friction units in various fields of technology (Zhang et al., 2005; Musil et al., 2010; Stuber et al., 2002).

Solution of Problem.

A promising area of research in this area are coatings based on amorphous carbon (a-C), used as a solid lubricant, and nanosized crystalline particles of carbide and / or) nitride phases. These coatings are of the type DNG/AM (dispersed nanograins in amorphous matrix, dispersed nanograins in an amorphous matrix) Musil (2012) and have several advantages over diamond like carbon (DLC), which have high internal stresses and, therefore, low adhesion and fracture toughness, as well as MoS_2 coatings with low hardness and therefore low bearing capacity (Stuber et al., 2002; Teer, 2001; Fox et al., 2000).

Coatings based on pure amorphous carbon (not containing additional elements) have a low coefficient of friction ($f < 0.1$), and a low coefficient of wear ($k \sim 10^{-7}$ mm^3 N^{-1} m^{-1} and high hardness ($H \leq 24$ GPa) (Martinez-Martinez et al., 2009; Musil et al., 2010; Zhang et al., 2004; Tay et al., 2001)[14, 21, 25, 26]. However, their significant drawback is the presence of high internal stresses that limit both the thickness of the resulting coatings and the possibility of their use, due to the formation of cracks under high loads The physical cause of high internal stresses is a significant fraction of diamond-like bonds of sp ^ 3 type in amorphous carbon (Lei et al., 2016; Musil et al., 2010; Zhang et al., 2004; Tay et al., 2001). A significant modification of both the microstructure and the nature of the interatomic interaction with the exception of this drawback is primarily associated with solid-solution doping of amorphous carbon, for example, using *Cr, Si*, *Al*, *Ti* (Musil, 2012; Musil et al., 2010; Zhang et al., 2004; Tay et al., 2001). In this case, the proportion of diamond-like bonds of sp^3 type decreases in amorphous carbon and the proportion of graphite-like bonds of sp^2 type increases which leads to a significant decrease in internal stresses, however, the hardness of the coatings may decrease (Musil, 2012; Musil et al., 2010; Zhang et al., 2004; Tay et al., 2001).

Apparently, the hardnessH, the cohesive and adhesive properties of nanocomposite coatings, in addition to the above internal stresses and grain size, should also be determined by the subtle features of the defective structure of nanocrystals and their boundaries, the presence of impurities (in particular argon) when using the magnetron method of their preparation, the nature distribution of atoms of the main elements of the composition in the volume of coatings. These issues are still poorly studied in connection with the experimental difficulties of research in nanocrystalline materials.

The presence of amorphous carbon nanoscale particles of strengthening phases (carbides, nitrides, etc.), which are introduced or synthesized during the deposition process, can effectively increase the hardness H of the resulting nanocomposites. By changing the ratio of volume fractions of the amorphous and nanocrystalline phases, one can control the mechanical and tribological characteristics (H, f, k, etc.). It was shown in Martinez-Martinez et al. (2009) that for *TiC/(a-C)* coatings, there is an optimal C/Ti ratio in the range of 3–9, which is determined by the number of graphite targets and the magnetron power. The specified ratio provides high values of H and low values of f and k.

An equally important characteristic of the coatings under discussion is the ratio of the hardness H to the effective modulus of elasticity of the coating $E^* = E/(1-v^2)$ (ν – Poisson's coefficient, E – Young's modulus), by changing which the adhesive properties can be controlled. Reducing the difference between the elastic moduli of the substrate and the coating allows one to increase the adhesive properties (Musil, 2012; Musil et al., 2010), while the value of the elastic modulus E^* changes only due to a change in the composition of the coating. For example, the introduction of a metallic phase with a relatively low modulus of elasticity (the soft phase – Cu, Al, etc.) into the coating increases the adhesion, but the hardness decreases (Tay et al., 2001). The ratio H^3/E^{*2} characterizes the resistance of the coating to plastic deformation, and also determines the coefficients of friction and wear (Janahmadov, 2020; Musil et al., 2010). To obtain coatings with values of $H < 20GPa$, $f < 0.1$, $k \leq 2 \cdot 10^{-7}\ mm^3\ N^{-1}\ m^{-1}$, it is necessary to meet the conditions of the transition contact criterion (Janahmadov, 2020):

$$H/E^* > 0.1\text{) and } H^3/E^{*2} = 0.15\text{-}0.30$$

Recent studies of the aforementioned tribological phenomena confirm our earlier results (Janahmadov, 1996, Janahmadov & Javadov, 2016), which show that tribological surfaces must have a certain combination of hardness and elasticity indicators, both for the substrate and for coatings, so that deformation under load occurs without elasticity. However, it should be noted that during normal contact interaction (without coating), when temperature-force factors act on the surface, then instead of hardness it is better to use the voltage value for the ratios, i.e. σ/E (Janahmadov, 1996).

As studies Musil, (2012) show, the introduction of Cr and Si in the coating, released at the grain boundaries, suppresses the grain-boundary diffusion of oxygen, significantly increasing the corrosion properties of the coating, and the presence of these elements in a solid solution reduces the internal tension in the amorphous matrix (Musil, 2012; Teer, 2001; Fox et al., 2000). The presence of nanosized particles in the amorphous matrix, such as titano-carbide — *TiC*, titanium nitride — *TiNi*, prevents of formation of the crack nuclei on them, thereby preventing their propagation (Musil, 2012). This increases cohesive and adhesive strength while maintaining high hardness. Thus, for the simultaneous implementation of a whole range of mechanical properties (high hardness, low friction coefficient, high adhesion, etc.), a fundamentally different approach to the design of coatings, including coatings based on amorphous carbon, is required. This approach is implemented on the basis of the principle of multi-element (Коротаев et al., 2011; Janahmadov, 2020), which allows controlling a number of characteristics. In this regard, the choice of the elemental composition of the coatings is determined by physical considerations of the influence of specific elements of metallic and nonmetallic (carbide, nitride) phases on the required set of mechanical properties.

In Андреев et al. (2015), the method of the electron microscopy transmission is used to study the microstructure and properties of multi-element nanocomposite coatings based on amorphous carbon and nanosized particles using the TiCNiCr/(a-C) coating as an example. The results of the study confirmed the presence of nanocrystalline (TiC and NiCr) phases and an amorphous (a-C) phase in the coating. Figure 1 Андреев et al. (2015) shows dark-field images of this coating and the corresponding diffraction patterns.

In the diffraction patterns, a diffuse halo corresponding to the amorphous phase, as well as diffraction rings corresponding to the TiC crystalline phase with a small crystallite size, and brighter reflections corresponding to relatively large NiCr particles are observed. In regions with a high density of crystalline phases, the diffuse halo is weak (Fig. 1b); in areas with a high density of amorphous phase (Fig. 1d), a predominantly diffuse halo is observed, and the intensity of reflections of crystalline phases is very low. It is known that impurity metal atoms can dissolve in amorphous carbon, in particular chromium up to 20 at. % (Teer, 2001; Fox et al., 2000). Therefore, we can assume that in the composite coating under study, chromium partially dissolves in the amorphous phase; in the present coating, chromium partially dissolves in the amorphous phase.

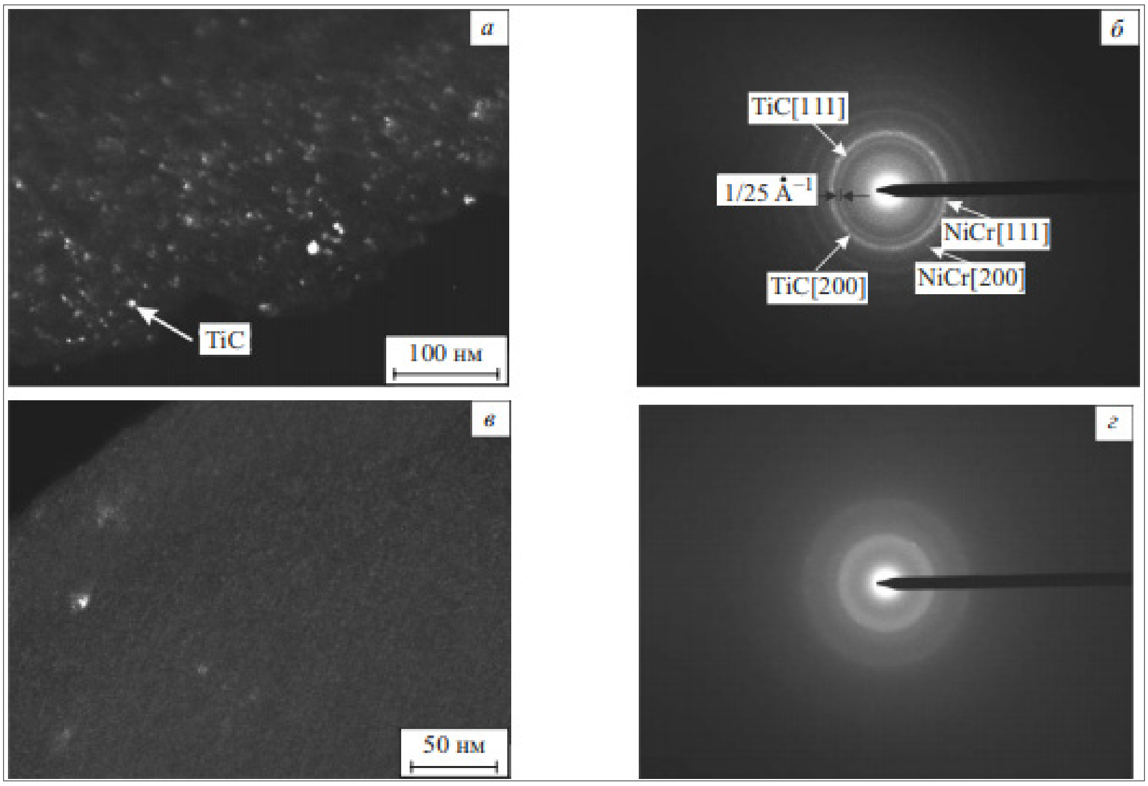


Figure 1: *TiCNiCr/a-C* coating: dark-field image in the [200] TiC reflection (a); dark-field image in an amorphous carbon ring (c); corresponding diffraction patterns (b, d). Reflections of the TiC and NiCr phases are noted. Transmission Electron Microscopy (Андреев et al., 2015)

Particles of titanium carbide are distributed in the amorphous matrix quite uniformly. Areas with a high density are observed (Fig. 1), when the distances between the particles are comparable with their sizes, and the areas where the distances between the particles significantly exceed these sizes. In the diffraction pattern (Fig. 1b), the rings from the TiC phase are broadened, which indicates a small crystallite size. Estimates of the sizes at the half-width of the diffraction maximum [100], which is 1⁄25 A^{-1} for the reflection = [111] *TiC*, give values of ≈2.5*nm*, which is consistent with the data of dark-field images.

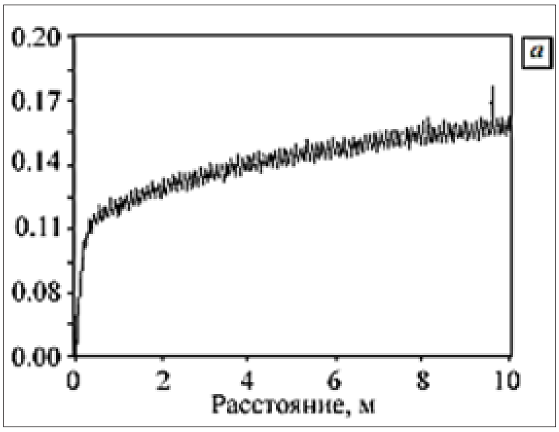
In addition to nanoscale *TiC* particles, relatively large (up to 30nm) particles (or conglomerates of particles) with interplanar spacings close to *Ni* (*NiCr*) were found. As indicated above, these particles can be a solid solution of chromium (up to 20*at.*%) in nickel. In the diffraction patterns, such particles (conglomerates of particles) give separate brighter reflections (Fig. 1, b, d).

It is known that the dark-field analysis with successive tilting of the foil in the goniometer makes it possible to identify local features of the elastically stressed state of the material by evaluating the components of the curvature tensor – the torsion of the crystal lattice.

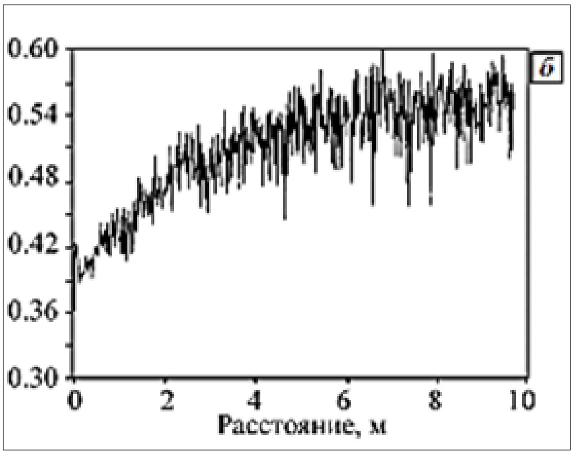
Studies of the mechanical properties of *TiCNiC*r / (*a-C*) coated titanium alloy samples showed that the microhardness of such a composition is approximately 7 times higher than that of a titanium alloy. The hardness of the coating based on amorphous carbon with titanium carbide nanoparticles was ≈14*GPa*, with the initial hardness of the titanium substrate 2*GPa*. In this case, at a coating thickness of ~2.8*μm*, we are talking about the microhardness of the substrate – coating composition, since the indenter penetration depth in the process of microhardness measurement exceeds 0.1 of the coating thickness.

The study of the tribological characteristics of the samples also showed significant changes in properties due to the physical nature and structure of the coating. In Fig. 2 Андреев et al. (2015) presents the dependences of the friction coefficient on the distance traveled by the indenter for a sample with and without coating. From the analysis of these dependencies it follows that the value of the coefficient of friction for the studied coating (up to 10*m*) was f≈0.14-0.16. The study of the surface profile of the coating indicates a relatively high (R_ z~0.35*μm*) roughness. An increase in power (up to 1.5*kW*) at the composite cathode negatively affects both the roughness and the structure of the coatings; the roughness of the coating increases and large particles (conglomerates of particles) of *NiCr* with sizes up to 100nm are observed in the coating. The presence of these particles also leads to a certain decrease (up to 11*GPa*) of the microhardness of titanium coated samples.

From a comparison of the dependencies in Fig. 2 a and b that the coefficient of friction for a friction pair of titanium alloy is very high ($f \approx 0.5$-0.7) (Fig. 2b). These circumstances impede the effective use of titanium products in friction units, including those intended for work in aerospace vehicles. Nanocomposite coatings of the *Ti–C–Ni–Cr* system based on amorphous carbon on titanium products can significantly reduce the friction coefficient (to $f \approx 0.14$-0.16) and significantly increase the wear resistance of titanium products (lightweight gears, drives, etc.). The physical reason that reduces the coefficient of friction is the structure of amorphous carbon, which plays the role of a solid lubricant under friction conditions.



Distance, m



Distance, m

Figure 2: Dependence of the coefficient of friction on the path length of the indenter: a) a sample of a titanium alloy coated with TiCNiCr ⁄ a-C; b) an uncoated titanium alloy sample.

The increase in microhardness of the composition “titanium substrate – coating *TiCNiCr / a-C*” (approximately 7 times relative to the titanium alloy) is of significant practical importance for the relative soft titanium alloys. Such an increase is provided by the *DN6 / AM* coating structure containing an amorphous phase having a sufficiently high hardness due to the presence of diamond-like bonds типа sp^3 in it and also hardening nanosized particles of the carbide phase *TiC*. Nanosized particles of *NiCr*, apparently, also contribute to the increase in hard coatings. Based on the analysis of the above literature sources, it can be assumed that the presence of *Cr* in the solid solution of amorphous carbon, and possibly *Ni,* leads to an increase in graphite-like bonds (the sp^2 type) and a decrease in diamond-like bonds of the sp^3 type. Such changes in the structure of amorphous carbon provide a sufficiently high value of microhardness H, a low level of internal stresses, and low values of the effective Young’s modulus E^*, which makes it possible to obtain a coating with a thickness of several microns with sufficiently low internal stresses, good adhesive properties, and ductility.

Conclusion

The optimal choice of the ratio of hardness to effective elastic modulus for a certain combination of substrate and coating, taking into account their tribological properties, is established, which is the most important characteristic of nanostructured coatings and is implemented on the basis of the multielement principle.

Nanocomposite coatings based on amorphous carbon on titanium products provides high values of hardness and low values of the coefficient of friction due to the presence of diamond-like bonds in them, and as a result, the wear resistance of the product significantly increases. This makes it possible to use them in friction units, especially in aerospace machines, mechanisms and devices.

An equally important characteristic is the ratio of hardness to the effective modulus of elasticity of the coating. A criterion for transient contact conditions $J_k = H/E$ is introduced, which is the ratio of hardness to elastic modulus.

Reducing the difference between the elastic moduli of the substrate and the coating allows to increase the adhesive properties by changing the composition of the coating with the amorphous phase contained in it.

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