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Research article

Surface modification of coatings based on Ni-Cr-Al by pulsed plasma treatment

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Abstract: To protect materials from abrasion-corrosion, various thermal spraying methods can be used to apply coatings, such as gas-flame powder spraying, plasma spraying, high velocity oxygen-fuel spraying and detonation cannon. Thermal spraying is one of the most effective methods of protecting the material from wear and corrosion, thereby increasing the service life of the material used. We present the surface modification of coatings based on Ni-Cr-Al by a pulsed plasma treatment using a plasma generator. The coatings were obtained by detonation spraying followed by pulsed plasma treatment. The changes to the structural properties of the coatings under the influence of plasma flow were studied using scanning electron microscopy, energy-dispersive X-ray spectroscopy and X-ray diffraction. The mechanical and tribological properties were analyzed by surface roughness characterization, microhardness testing and tribological tests for a comprehensive analysis of changes in the characteristics of the Ni-Cr-Al coatings following pulsed plasma treatment. It was found that modification of the coating by pulsed plasma treatment causes an increase in the microhardness of the surface layer, as well as a reduction in the surface roughness and friction coefficient. According to the results of X-ray phase analysis, these improvements in the mechanical and tribomechanical properties of the obtained surfaces is associated with an increase in the content of $CrNi_3$, NiAl and NiCr phases in the coatings.

Keywords: thermal spraying; detonation gun; pulsed plasma treatment; surface coating; friction coefficient; roughness

1. Introduction

Coatings are used in a variety of applications including automotive systems, boiler components and power equipment, chemical process equipment, aircraft engines, orthopaedics and dentistry, land and marine turbines and ships [1]. Among the commercially available thermal spray coating methods, detonation spraying (DS) and high velocity oxygen fuel (HVOF) spraying are the best choices for obtaining the desired hard, dense and wear resistant coatings [2].

The required coating properties can be achieved using various methods, one of which is thermal spraying. Thermal spraying refers to the deposition of a material on a substrate in the form of softened or melted droplets to form a continuous coating. It is an efficient and inexpensive coating method for changing the surface properties of a material and has become an important tool in surface technology [3]. There are many thermal spray techniques that are used to apply coatings, such as flame powder spraying [4], arc wire spraying [5], plasma spraying [6,7], high velocity oxy-fuel spraying (HVOF) [8] and detonation gun spraying [9]. The choice of process depends on the functional requirements, the adaptability of the coating material to the intended method, the level of adhesion required, the availability of the method and the cost of the equipment [10].

Detonation-sprayed coatings play an important role in protecting materials and alloys from wear and corrosion phenomena [11]. Various researchers have carried out work on the study of the characteristics of coatings formed by detonation spraying [12,13]. However, there are still few works in the field of using nanostructured powders with detonation spraying. Additional research is also needed to optimize the technological parameters of the detonation spraying process, to improve the design of the detonation gun spraying device [14] and to understand how the technological parameters of detonation spraying affect the microstructure, mechanical and other properties of coatings [15].

The detonation gun separation device developed by researchers has led to good detonation gun atomization performance under high performance requirements [16]. The detonation gun spray process used in this work is similar to that described by authors [17]. The coating material is fed through a dosing vessel in the form of powder at the same time as a mixture of combustion gases (e.g., oxygen and acetylene). Subsequently, a spark plug ignites the gas inside the chamber, generating a detonation wave that propels the coating material through a metal barrel towards the substrate at very high velocities. The resulting high kinetic energy of the hot powder particles upon impact with the substrate leads to the formation of a very hard, dense and durable coating. The coating thickness depends on various process parameters, including but not limited to, the particle size of the coating material, the ratio of combustion gases used, the gas flow rate and the distance between the barrel outlet and the substrate. The detonation spray cycle can be repeated at a rate of 1–10 rounds per second until the desired coating thickness is obtained [18].

The quality of coatings obtained by detonation spraying depends on two major parameters: the velocity V_p and the temperature T_p of the particles upon collision with the surface. During detonation spraying, the optimal conditions for the formation of the coating correspond to the semi-molten state ($T_p = T_s$) of the sprayed particles (spraying temperature, T_s) before colliding with the substrate surface. Particles that have not reached the semi-molten state are in most cases repelled from the surface, thereby reducing the deposition rate. Otherwise, if such particles are embedded in the layer, they reduce the quality of the coating, causing the formation of defects and irregularities. Overheating of the particles is undesirable, since this leads to the splashing of drops, which also reduces the quality of the surface of the resulting coating [19].

Preliminary results have shown that the phase composition of detonation coatings can vary significantly depending on the thermal annealing temperature. However, such heat treatment has disadvantages related to the softening of the substrate materials. The disadvantages of traditional heat treatment methods can be overcome by thermal activation of the surface by pulsed plasma flows [20]. The advantages of pulsed plasma treatment (PPT) are the high rates of heating and cooling of the material surface (10^4–10^8 Ks^{-1}), as well as the possibility of creating layered structures with different phase compositions and, accordingly, with different physicochemical characteristics. There is also the possibility of local impact on the product by pulsed plasma. Thus, the pulse-plasma technology makes it possible to control the surface layer in the process of change. There is still no general opinion in the literature about the nature of the effect of PPT on structural-phase transformations in coatings based on Ni-Cr-Al, or how the distance from the plasma torch to the sample surface influences the mechanical and tribological properties of coatings. In this article we investigate the effect of processing distance during PPT modification of Ni-Cr-Al coatings obtained by detonation spraying, under the influence of a plasma flow, on the mechanical and tribological properties of the resulting surfaces.

2. Materials and methods

Coatings based on Ni-Cr-Al were obtained using the detonation installation "Computer Controlled Detonation Spraying (CCDS2000)" (Research Centre "Surface Engineering and Tribology", Ust-Kamenogorsk, Kazakhstan), in which detonation is carried out inside the barrel in an explosive mixture, formed as a result of the flowing supply of gas components through a specialized mixing device (see Figure 1a) [21].

The PPT surface modification of the coatings was carried out by a pulsed plasma treatment using a plasma generator developed by the Institute of Electric Welding named after E.O. Paton of the National Academy of Sciences of Ukraine. A feature of this plasma generator is the possibility of switching the electric current by the ionized gas region behind the front of the detonation wave. This makes it possible to generate pulsed plasma with a frequency of 1–4 Hz and an energy of up to 7 kJ (Figure 1b).

The substrates were grit blasted prior to spraying to increase the surface roughness and therefore improve adhesion (Substrate: DIN 14MoV63 steel, arithmetic mean deviation of the profile R_a = 1.25 µm). Composite powder 584.054 produced by H.C. Starck was used as sprayed material. The feedstock was a mixture of 80 wt.% NiCr powder (Ni20Cr80) and 20 wt.% Al (99.99% purity). The nominal particle size of the powder varied from 30 to 45 µm. The fuel gas used was an acetylene-oxygen mixture, which is the most commonly used fuel for the detonation of sprayed powder materials. The spraying was carried out at the ratio of acetylene-oxygen mixture O_2/C_2H_2 = 1.856. The volume of the explosive mixture of the detonation pistol barrel was 60%. Nitrogen was used as the carrier gas. The distance between the treated sample surface and the detonation barrel was 250 mm. Detonation coatings were treated with a direct-action pulsed plasma arc in the following modes: capacitor capacitance 960 µF, voltage 3.2 kV, inductance 30 µF, frequency 1.2 Hz, passage speed 3.4 mm/s, electrode recess h = 10 mm, number of passes 1. Processing modes were selected by changing the distance H from the plasma torch to the hardened surface (see Table 1).

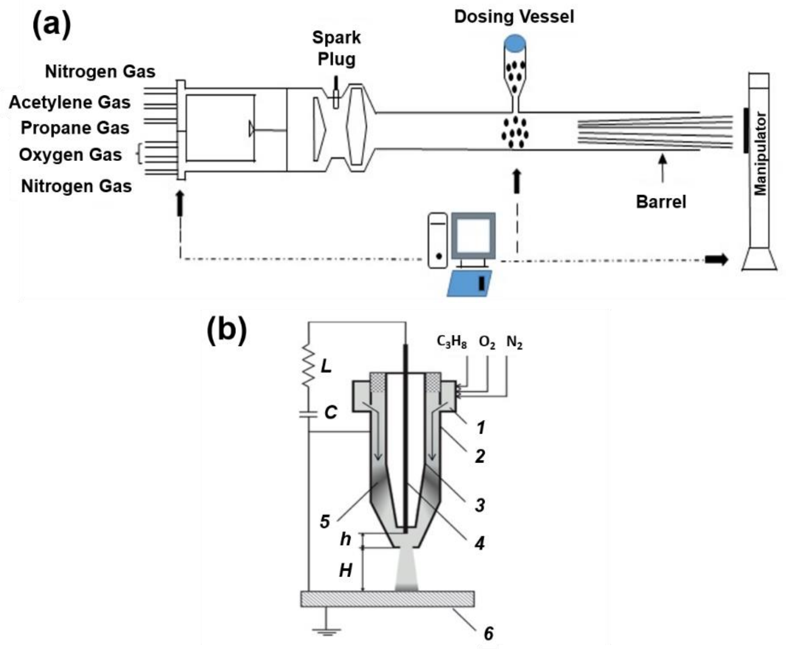

Figure 1. Principal diagram of a detonation complex "CCDS2000" (a) and PPT (b):1—detonation chamber; 2,3—coaxial electrodes; 4—consumable electrode; 5—detonation wave; 6—treating part [21].

Table 1. Technological parameters of PPT processing by the method of Ni-Cr-Al coatings.

Sample No.	Coating	Distance from the plasma torch to the sample surface, H, mm	Average roughness (R_a), µm	Hardness, GPa
1 (Initial)		-	9.83	4.1
2	Ni-Cr-Al	40	4.47	7.4
3		50	6.12	6.2
4		60	3.82	7.6

The chemical compositions and morphology of the coating were examined using X-ray energy-dispersive spectrometer system "OXFORD INSTRUMENTS", by scanning electron microscopy and energy-dispersive X-ray spectroscopy (SEM/EDX) "VEGA3 TESCAN", "TESCAN SEM Solutions" and "NeoScope JEOL JSM-5000" with voltages of 10–20 kV.

To study the crystal structure of the deposited coatings, X-ray diffraction (XRD) was performed using an X-ray diffractometer "D8 DISCOVER" (Bruker) at 40 kV and 40 mA with a copper target (λ = 154 nm). The scan had a step size of 0.02° and started from 10 to 90°. A multifunctional adapter was installed to carry out grazing angle of incidence diffraction.

To analyze the surface roughness R_a, as well as the volume of the removed coating material, we determined the section of the wear track on the surface of the sample using the "Model 130, JSC Plant Proton" (contact) and "Alicona InfiniteFocus Model G5, Bruker" (not contact) profilometers (ASTM D7127-05).

Microhardness measurements were carried out on an automated hardness tester of the model "METOLAB" using the Vickers method with an indenter load of 0.2 N (ASTM E384-11 covers Vickers hardness testing using forces in the range 9.807×10^{-3} to 9.807 N). The microindentations were made at a distance of 1.0 mm from each other.

Tribological tests were carried out in air at a room temperature of 28 °C according to the "ball-disk" scheme on a "Tribometer TRB-3" (Anton Paar) friction machine. A ball with a diameter of 3.0 mm, made of sintered certified material-SHKh15, was used as a counter body. The load was 6.0 N, the sliding speed was 2.0 cm/s, the track radius was 2.5 mm and the total travelled path of the counter body was 50 m. The test conditions were in accordance with the international standards ASTM G99 and ASTM G133-95. The friction coefficients reported are the mean value across the sliding distance (50 m).

3. Results and discussion

According to the results of scanning electron microscopy, detonation spraying and subsequent pulsed plasma treatment have several features in the formation of surface morphology. Figure 2a shows an SEM image of the Ni-Cr-Al surface before PPT. The coating has a typical structure characteristic of thermal spraying methods: high density, uniform composition and layer thickness, as well as the presence of individual pores. Figure 2b–d show SEM images of the Ni-Cr-Al surface after PPT. It can be seen that the surface layer contains a melted plasma-modified complex-gradient layer. Prior to PPT, the surface coating displays significant spatial variation in the elemental composition. Across the four sites at which EDX was performed for sample 1, the atomic percentage of Ni ranges between 15.49% and 64.82%, the atomic percentage of Cr ranges between 8.79% and 19.17% and the atomic percentage of Al ranges between 8.74% and 70.10%. As a result of pulsed plasma treatment, the chemical composition of the surface of the Ni-Cr-Al coatings changes depending on the distance to the plasma torch with a change in the intensity of individual elements such as Al, Ni, Cr and Fe (see Table 2 and Figure 3).

This agrees well with the mean ratio of Ni to Cr measured by EDX (averaged at four sites) being approximately 2.9 to 1. The emergence of the Fe peak at 6.4 keV in the EDX spectra (Figure 4) reveals that the coating surface increasingly contains Fe as the distance H between the plasma torch and the sample surface during PPT is reduced. When this distance H is 40 mm, the atomic percentage of Fe measured by EDX is as high as 69.97%, compared to just 2.48% prior to PPT. The atomic percentage of Fe measured by EDX is still present but less significant for sample 3 and 4, when H is equal to 50 and 60 mm, respectively. It is plausible that in the reaction zone, where the temperature can reach 3500 °C, complete melting of the deposited coating and partial melting of the Fe containing substrate occurs. The Al_2O_3 phase is significantly more prevalent after PPT treatment, which is performed in ambient conditions, i.e., in the presence of oxygen.

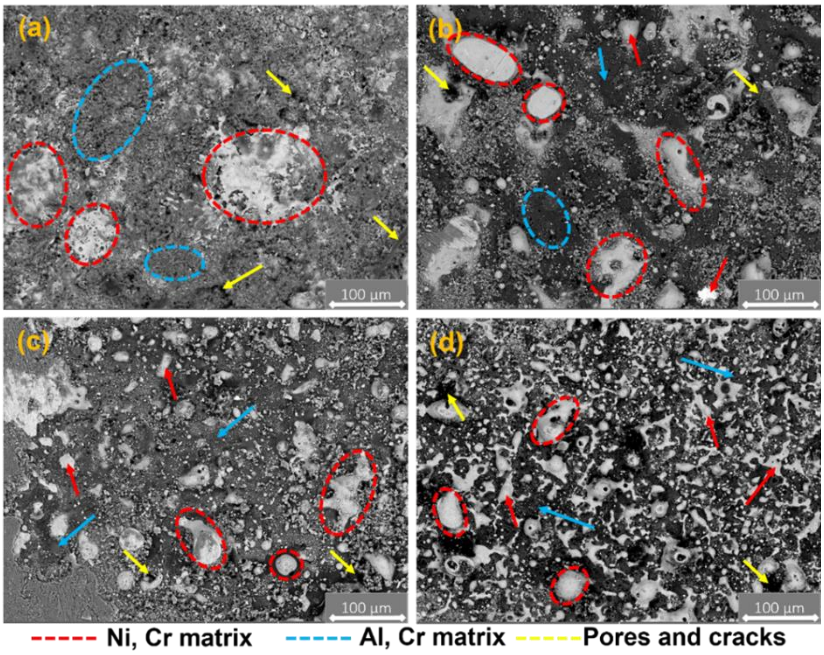

Figure 2. SEM-image of the surface of Ni-Cr-Al coatings before (a) and after (b), (c) and (d) PPT.

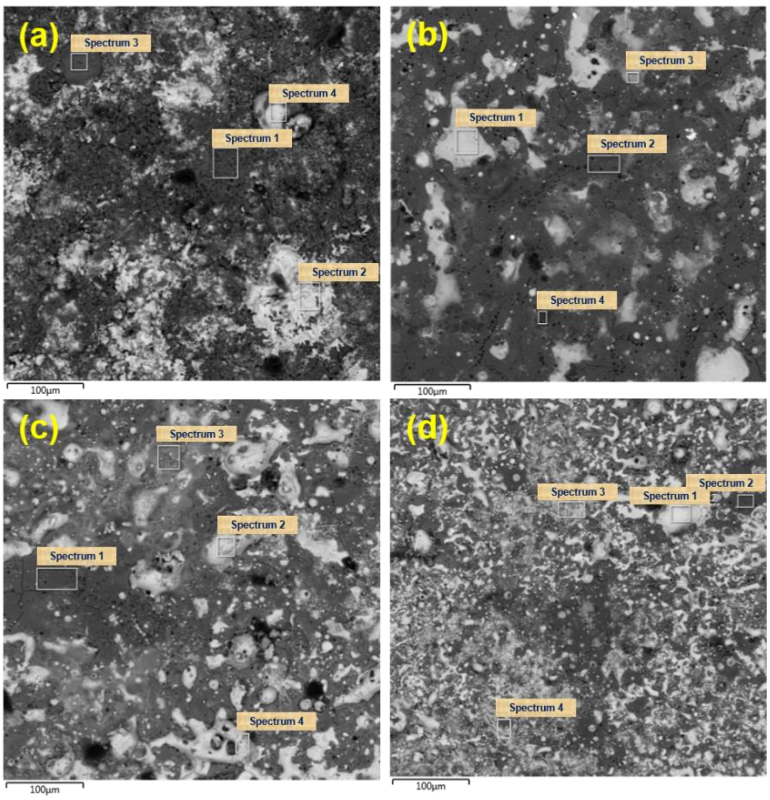

Figure 3. SEM-image of the surface spectrum areas of Ni-Cr-Al coatings before (a) and after (b), (c) and (d) PPT (phase contrast).

Table 2. SEM/EDX chemical composition of elements in the Ni-Cr-Al coatings.

Sample No.	Spectrum areas	Elemental composition, at. %						
		Ni	Cr	Al	Fe	O	C	Impurity
1	Spectrum 1	34.46	10.91	50.70	0.52	2.46	0.72	0.23
	Spectrum 2	64.82	19.17	8.74	2.48	3.17	1.38	0.24
	Spectrum 3	15.49	8.79	70.10	0.48	8.79	1.02	0.29
	Spectrum 4	52.90	16.01	13.37	0.90	6.25	10.07	0.28
2	Spectrum 1	9.81	4.23	1.05	69.97	8.64	5.46	0.83
	Spectrum 2	17.74	3.23	32.14	7.18	38.76	0.61	0.35
	Spectrum 3	7.58	3.65	1.35	49.29	18.20	19.32	0.61
	Spectrum 4	39.13	0.50	2.30	32.60	24.52	0.45	0.29
3	Spectrum 1	42.72	10.59	38.24	2.86	3.83	1.39	0.37
	Spectrum 2	44.64	15.13	7.02	18.01	10.31	1.95	0.38
	Spectrum 3	6.61	14.76	35.29	20.80	14.92	6.85	0.42
	Spectrum 4	26.54	2.93	47.66	8.05	11.95	2.43	0.10
4	Spectrum 1	62.15	17.42	1.67	7.03	5.21	4.69	1.84
	Spectrum 2	16.05	37.94	39.60	2.25	2.40	1.11	0.64
	Spectrum 3	31.47	8.35	10.19	25.96	15.15	7.39	1.49
	Spectrum 4	43.59	13.80	16.06	5.18	13.41	2.55	1.84

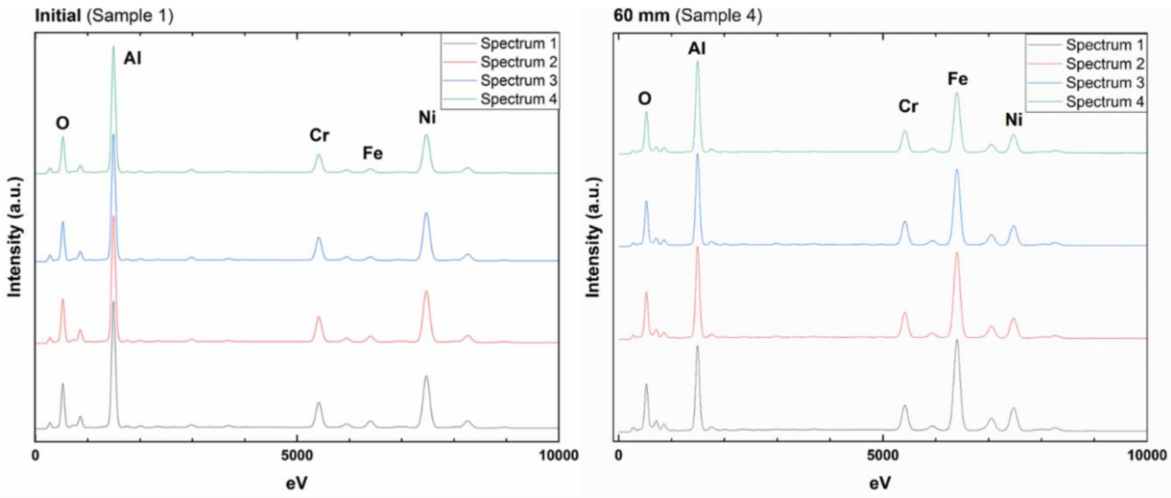

Figure 4. EDX-analysis of the surface of coatings based on Ni-Cr-Al elements.

XRD analysis was performed on the surface of the resulting coatings. The depth of probing or penetration of the X-ray beam is approximately 0.3 µm, depending on the angle of incidence of the beam. The presence of an Al_2O_3 peak, detected only in the treated coatings, can clearly be attributed to pulsed plasma treatment in an oxygen-containing atmosphere. Phase analysis of these coatings by XRD indicates the presence of $CrNi_3$, NiAl, NiCr, Al and Al_2O_3 phases (see Figure 5). Most likely, during sputtering by the detonation method and subsequent PPT, the substrate surface is heated to melting and the $CrNi_3$, NiAl and NiCr phases are formed from the deposited Ni, Cr and Al elements. Prior to PPT, the deposited Ni-Cr-Al coatings appear to consist primarily of $CrNi_3$ with some aluminium-containing phases also present.

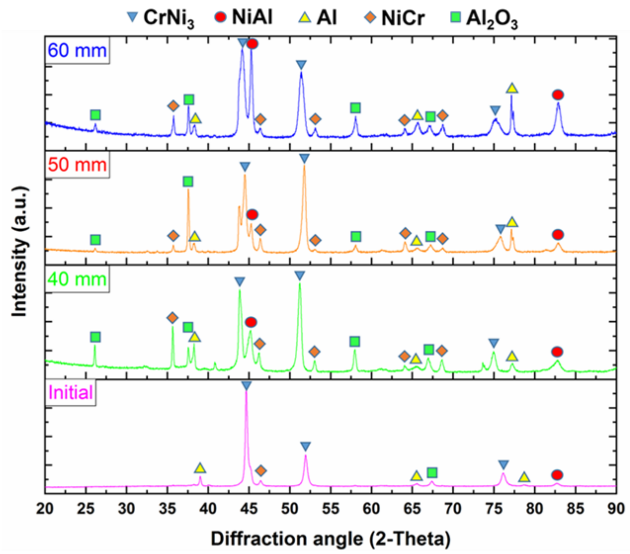

Figure 5. XRD-diffraction patterns of the Ni-Cr-Al coatings sprayed by the DS (initial) and after PPT (40, 50 and 60 mm).

Figure 6 shows 3D images of the surface of the Ni-Cr-Al coatings before (initial) and after (40, 50 and 60 mm) PPT obtained by profilometry. It can be seen that the surface morphology is affected by PPT. In particular, the surface roughness is significantly decreased (see Table 1); PPT smoothes the surface and reduces irregularities. The mean deviation R_a before PPT was 9.83 µm and the mean deviation after PPT (for the largest distance, 60 mm, between the surface and the plasma torch) was 3.82 µm, a decrease of approximately 39%. This is consistent with previous reports in the literature [22].

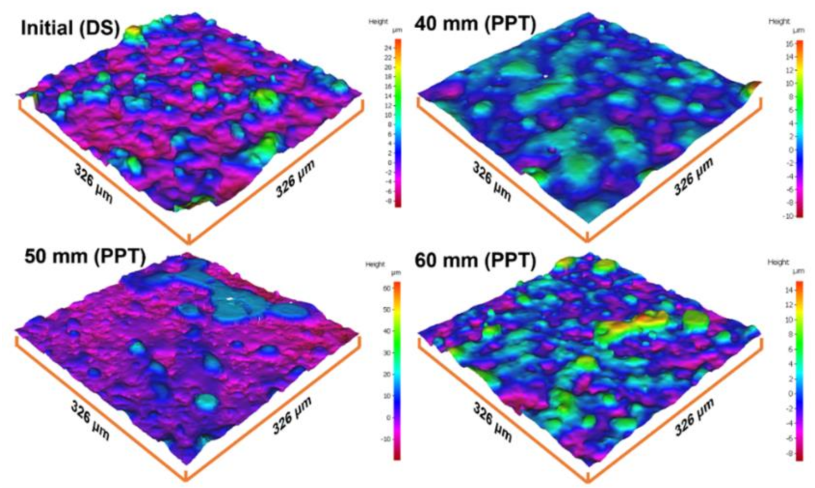

Figure 6. 3D-colour map images of the surface roughness of Ni-Cr-Al coatings before (initial) and after (40, 50 and 60 mm) PPT, when magnifying the eyepiece of an optical microscope by 50x.

One of the most universal parameters that allow one to quickly evaluate the mechanical properties of the obtained coating is the determination of its microhardness [23,24]. The average microhardness of the Ni-Cr-Al coatings before PPT (initial) was 4.1 GPa and after PPT (40, 50 and 60 mm) 7.4, 6.2 and 7.6 GPa, respectively (see Table 1). The maximum hardness of 7.6 GPa is achieved at a distance H of 60 mm between the plasma torch and the sample surface. Increased hardness and wear resistance are associated with the formation of an alloy of the NiAl phase. This compound has such features as a relatively high melting point and high resistance to corrosion and oxidation [25].

Figure 7 shows the friction curves of coated Ni-Cr-Al before and after modification by PPT, depending on the distance H between the surface and the plasma torch. The experiment showed that after PPT the friction coefficient decreases, the mean value of the friction coefficient along the sliding distance of 50 m in Ni-Cr-Al coatings without treatment is 0.545. After PPT treatment with H = 40 mm, the mean friction coefficient is increased slightly to 0.574. However, when H is increased to 50 and 60 mm during PPT, the mean friction coefficient decreased to 0.480 and 0.415, respectively. These results clearly show that the friction coefficient is decreased as the distance H between the surface and the plasma torch is increased. This is consistent with the measured surface roughness of the surfaces: the smallest friction coefficient corresponds to the smallest R_a (see Table 1). The experiments performed have shown that PPT leads to an improvement in the tribological properties of Ni-Cr-Al coatings. This effect can be obtained by eliminating surface defects (microcracks and pores) and changing the structural-phase state of coatings. In the process of friction, several characteristics simultaneously affect the surface, therefore the coefficient of friction is not strictly determined by the hardness values. However, one of the possible reasons for the decrease in the friction coefficient may be an increase in microhardness. According to reference [25], surface hardening and a decrease in the friction coefficient have a direct correlation.

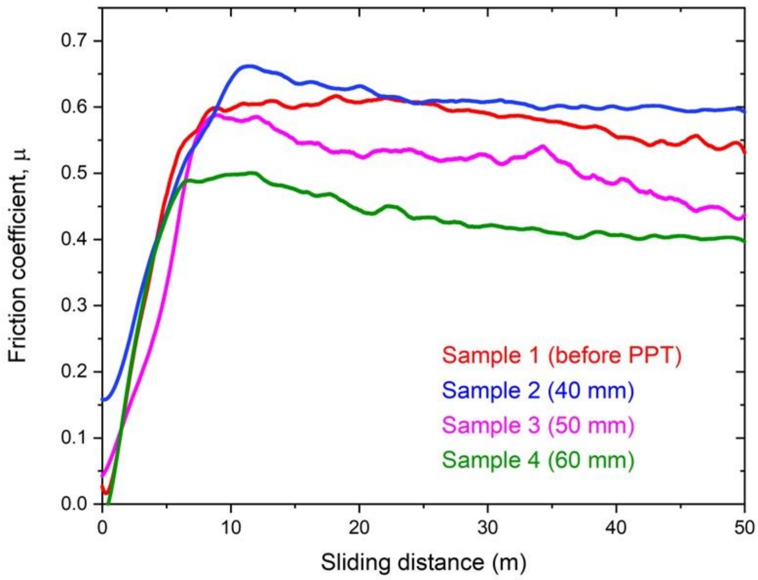

Figure 7. Results of tribological tests of Ni-Cr-Al coatings before and after PPT. The data presented has been smoothed with a Savitzky-Golay filter (using a 4th order polynomial and 3000 data points).

It is known [26] that the hardness of metal matrix coatings depends on matrix grain refinement and on precipitation hardening, which was in this work achieved by PPT. According to the data obtained, it was revealed that the sample with the lowest surface roughness and friction coefficient also had the highest value of microhardness (see Table 1 and Figure 6).

4. Conclusions

In this work, the influence of the pulsed-plasma treatment distance during the surface modification of coatings based on Ni-Cr-Al obtained by the detonation method was studied. Pulsed-plasma treatment was found to reduce the surface roughness and reduce irregularities regardless of the processing distance. The surface roughness R_a after pulsed-plasma treatment was approximately 39% lower after pulsed-plasma treatment. The experiment performed in this work show that after pulsed-plasma treatment, the friction coefficient decreases from 0.545 to 0.415 (for the largest treatment distance). The average microhardness of the Ni-Cr-Al coatings before pulsed-plasma treatment was 4.1 GPa and after treatment was increased by a factor of 1.5–1.8, depending on the distance between the between the surface and the plasma torch. Therefore, we have demonstrated that pulsed-plasma treatment can be used for improving the mechanical and tribological properties surface of coatings obtained by the detonation gun spraying method, and that a larger distance between the surface and the plasma torch results in a harder surface coating.

Use of AI tools declaration

The authors declare they have not used Artificial Intelligence (AI) tools in the creation of this article.

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Conflict of interest

The authors declare that they have no conflict of interest.

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