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OPTIMIZING DEPOSITION PARAMETERS OF AlCrTiN COATINGS FOR IMPROVING THE WEAR RESISTANCE AT ELEVATED TEMPERATURES

OPTYMALIZACJA PARAMETRÓW OSADZANIA POWŁOK AlCrTiN DLA POPRAWY ICH ODPORNOŚCI NA ŚCIERANIE W PODWYŻSZONEJ TEMPERATURZE

Key words:

AlCrTiN coatings, cathodic arc deposition method, Taguchi method, coating mechanical and tribological properties

Słowa kluczowe:

powłoki AlCrTiN, łukowo-próżniowa metoda osadzania, metoda Taguchi, właściwości mechaniczne i tribologiczne

Summary

This article presents the investigation performed to develop process parameters to produce AlCrTiN coatings with improved mechanical properties and the best wear resistance using the cathodic arc evaporation technique. The chemical composition of targets, nitrogen pressure, arc current, and bias voltage with three levels of values were selected as important parameters of deposition that

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determine the mechanical and tribological properties of the AlCrTiN coatings. The Taguchi method was applied to study the impact of deposition process parameters on the coatings properties and for their optimization. Based on the statistical analyse, the most effective deposition parameters and optimum deposition conditions to obtain the best tribological behaviour were determined. The results showed, from among investigated parameters of deposition process, that the targeted chemical compositions, bias voltage, and nitrogen pressure have a significant effect on the mechanical properties and tribological behaviour of AlCrTiN coatings. It was also found that, to achieve both good mechanical properties and the best wear resistance of the AlCrTiN coatings, the deposition process by cathodic-arc technique should be carried out as follows: by using targets with composition Al70%Cr15%Ti15% and a negative bias voltage of 150 V, an arc current of 55 A, and a nitrogen pressure of 2.0 Pa.

INTRODUCTION

The ternary AlCrTiN hard coatings, deposited by PVD (Physical Vapour Deposition) methods, are a very promising choice for several industrial applications, such as cutting tools used for high performance machining of aerospace materials (Ni-based super-alloys, Ti-based alloys) **[L. 1]** and forging or punching tools **[L. 2]**. Previous studies have shown that these multicomponent coatings exhibit high hardness (25–35 GPa) **[L. 3–5]**, improved oxidation resistance at elevated temperatures (thermal stability to 900°C) **[L. 6]**, good adhesion strength to steel substrates **[L. 7]**, and excellent tribological behaviour **[L. 8]**. Among various physical vapour deposition methods for the creation of coatings, the cathodic arc evaporation technique is a very popular technique used in the industrial manufacture of tribological coatings **[L. 6–9]** due to following advantages: the high ionization degree of plasma, the high kinetic energy of the ions participating in the synthesis of the coating material, as well as the elevated deposition rate of the resulting coating. In this technique, properties of the coatings can be easily modifed by changing the basic technological process parameters, e.g., negative bias voltage, substrate temperature, arc current, reactive gas pressure, and chemical composition of the targets **[L. 3, 4, 10]**. In present paper, the Taguchi method **[L. 11–12]** was adopted to optimize selected cathodic arc deposition processes parameters for the best tribological properties of the AlCrTiN coatings.

EXPERIMENTAL DETAILS

Sample preparation

Steel discs with a diameter 25.4 mm and 6 mm thick made of hot working steel (1.2365), initially quenched and tempered to the hardness 490±10 HV and then nitrided to the hardness 960±20 HV were used as the substrates for the deposition of AlCrTiN coatings. All the substrates were prepared by mirror polishing (S_a = 0.02 μm) and ultrasonic cleaning in a multi-stage washer with a trichloroethylene bath. The commercial device MZ-383 Metaplas Ionon was used for deposition of coatings. Before deposition, the substrates were heated by using radiant heaters in the vacuum chamber at a pressure of 5.0×10^{-3} Pa to the temperature of 200°C. Next, the substrates were cleaned by a two-step etching process. The first step of this process was realized using an argon-ion etching process at a pressure of 5.0×10^{-1} Pa and a negative substrate bias voltage of 50–300 V. In a second step of etching, the substrate was biased with a voltage of –950 V and bombarded with Ti ions in a vacuum less than $5x10^{-3}$ Pa. All coatings were prepared in nitrogen by using the reactive cathodic-arc method, applying the four Al-Cr-Ti composite targets and three pure metallic Cr targets to achieve better adhesion of the AlCrTiN coating to the substrate, and a thin 0.1 μm intermediate chromium layer was deposited. To create the design of experiments (DOEs) for deposition of AlCrTiN coatings, four basic arc process parameters (i.e. targets composition, substrate bias voltage, arc current intensity, and nitrogen pressure) **[L. 13]** were chosen. For each process parameter, three levels of values were selected to control the mechanical and tribological properties of the coatings **(Tab. 1)**.

Table 1. Parameters and their levels selected for the performance of deposition processes of AlCrTiN coatings

Tabela 1. Parametry i ich wartości wytypowane do przeprowadzenia procesów osadzania powłok AlCrTiN

Parameters		Levels		
		1	2	3
Target composition (at. %)	T	$Al_{0.7}Cr_{0.15}Ti_{0.15}$	$Al_{0.7}Cr_{0.20}Ti_{0.10}$	$Al_{0.7}Cr_{0.25}Ti_{0.05}$
Bias voltage (V)	A	100	150	200
Arc current (A)	B	45	55	65
Nitrogen pressure (Pa)	C	1.0	2.0	3.0

The experimental plan for the deposition AlCrTiN coatings consisting of nine processes was developed by using a Taguchi orthogonal array L_9 **(Tab. 2)**.

Table 2. Design of experiments for the AlCrTiN coating deposition processes based on the Taguchi method

Tabela 2. Plan eksperymentów osadzania powłok AlCrTiN opracowany z wykorzystaniem metody Taguchi

Specimen no.	Parameters and levels				Parameter value			
					Target composition (% at.)	Bias voltage (V)	Arc current (A)	Nitrogen pressure (Pa)
S1	T_1	A_1	B_1	C_1	$Al_{0.7}Cr_{0.15}Ti_{0.15}$	–100	45	1.0
S2	T_1	A_2	B_2	C_2	$Al_{0.7}Cr_{0.15}Ti_{0.15}$	–150	55	2.0
S3	T_1	A_3	B_3	C_3	$Al_{0.7}Cr_{0.15}Ti_{0.15}$	–200	65	3.0
S4	T_2	A_1	B_2	C_3	$Al_{0.7}Cr_{0.20}Ti_{0.10}$	–100	55	3.0
S5	T_2	A_2	B_3	C_1	$Al_{0.7}Cr_{0.20}Ti_{0.10}$	–150	65	1.0
S6	T_2	A_3	B_1	C_2	$Al_{0.7}Cr_{0.20}Ti_{0.10}$	–200	45	2.0
S7	T_3	A_1	B_3	C_2	$Al_{0.7}Cr_{0.25}Ti_{0.05}$	–100	65	2.0
S8	T_3	A_2	B_1	C_3	$Al_{0.7}Cr_{0.25}Ti_{0.05}$	–150	45	3.0
S9	T_3	A_3	B_2	C_1	$Al_{0.7}Cr_{0.25}Ti_{0.05}$	–200	55	1.0

Characterization of AlCrTiN coatings

The quantified depth profile analysis of the elemental composition of AlCrTiN coatings were made by radio frequency glow discharge optical emission spectrometry (RF GDOES) using a GD-Profiler HR Jobin Yvon Horiba spectrometer. The coatings surface topography and roughness were investigated using an optical profilometer Talysurf CCI Taylor-Hobson. The hardness and elastic modulus of the coatings were measured with a CSM Nano-Hardness Tester. A Berkovich diamond tip was used at a controlled maximum depth mode. The maximum indentation depths in all measurements were approximately less than one-tenth of the coating thickness. The coating-to-substrate adhesion strength was evaluated by scratch testing with a CSM Revetest Instrument. The measurements are made using the following standard parameters: loading rate dL/dt = 100 N/min and scratching speed v = 10 mm/min. For the quantitative adhesion evaluation, minimal load at which specific damages of the coating occur in a form of a spalling was assumed as the critical failure load L_{C2}. The high temperature tribological tests of the AlCrTiN coatings were carried out in ambient air at a temperature of 500°C using a pin-on-disk high temperature tribometer (CSM Instruments). An unlubricated tribosystem was employed. All tests were conducted for a distance of 100 m, along a circular track of 10 mm diameter under the following conditions: a ceramic ball (Si_3N_4) with a 6 mm diameter was used as the counterpart, a normal load of 10 N, and a sliding speed of 0.1 m/s. For each specimen, three tests were repeated. To calculate the wear volume, the three profiles perpendicular to the sliding direction of the wear track (every 120°) were recorded by a Taylor Hobson profilometer. The wear volume W_v (in mm^3) was calculated by the multiplication of the average cross sectional area of the wear track, A, (calculated on the base of a Taylor Hobson profilometer measurements) by the sliding track perimeter. The calculation was performed according to Eq. (1).

$$W_v = A * \pi d \ [mm^3] \tag{1}$$

Where: A – average cross sectional area [mm^2] and d – wear track diameter [mm].

RESULT AND DISCUSSION

Surface morphology and chemical composition of AlCrTiN coatings

The thickness of all manufactured AlCrTiN coatings was 2.5±0.5 μm. The study of the surface coatings topography have showed that all inspected coatings possess numerous defects, i.e. microdroplets, pinholes, conical hillocks, and shallow craters, which significantly affect their surface roughness **(Tab. 3)**.

Table 3. Surface roughness of AlCrTiN coatings
Tabela 3. Chropowatość powierzchni powłok AlCrTiN

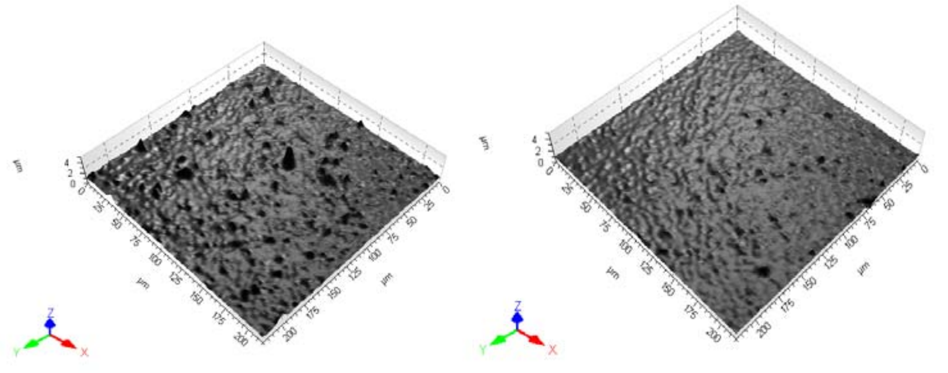
Specimen no.	S1	S2	S3	S4	S5	S6	S7	S8	S9
Average roughness S_a (μm)	0.20	0.08	0.07	0.07	0.11	0.08	0.07	0.12	0.13

The number of defects in the coatings mainly depends on the following two basic deposition process parameters: the bias voltage and the pressure. The microscopic study showed that the surfaces of coatings become smoother with the increasing of the values of these deposition process parameters, which significantly influence the value of the energy of ions involved in the synthesis and crystallization processes of coatings deposited by PVD methods **(Fig. 1)**.

The decreased quantity of defects in the coatings is caused by the following phenomena **[L. 13–14]**:

- A modification in the morphology of the coating induced by the ion bombardment mechanism, and
- An elimination of smallest microdroplets before they reach the surface of the coated substrate as a result their evaporation from the plasma stream by collisions with the ions, and
- A reduction emissions of microdroplets from targets of the arc sources due to increase of nitrogen pressure.

(a) (b)



Sa = 0.11 (μm) Sa = 0.07 (μm)

Fig. 1. Surface topography (3D images) and roughness of AlCrTiN coatings: a) S5 specimen (U_{bias} = –150 V, p = 1.0 Pa, I_{Arc} = 65 A; b) S3 specimen (U_{bias} = –200 V, p = 3.0 Pa, I_{Arc} = 65)

Rys. 1. Obrazy 3D topografii powierzchni oraz chropowatość powierzchni powłok AlCrTiN: a) na próbce S5 (U_{bias} = –150 V, p = 1,0 Pa, I_{Arc} = 65 A, b) na próbce S3 (U_{bias} = –200 V, p = 3,0 Pa, I_{Arc} = 65)

Based on the results of GDOES quantitative analysis, it was found that the chemical composition of the metallic components in the AlCrTiN coatings slightly differs in elemental composition from the targets used to manufacture the coatings **(Tab. 4)**. The observed deviations of the chemical composition between the coatings and the targets are caused by the following two characteristic phenomena of the cathodic-arc deposition process **[L. 10, 13, 15]**:

- Diversity in the evaporation rate of the targets components: Al, Cr, Ti; and,
- The selective re-sputtering of individual atoms from the growing coating (e.g., Al atoms) by a high-energy ion bombardment of the coatings surface during the deposition.

Table 4. The relative atomic ratio of metallic elements in the chemical composition of coatings AlCrTiN
Tabela 4. Względny udział atomowy pierwiastków metalicznych w składzie chemicznym powłok AlCrTiN

Specimen no.	Atomic ratio Al/(Al+Cr+Ti) (at.%)	Atomic ratio Cr/(Al+Cr+Ti) (at.%)	Atomic ratio Ti/(Al+Cr+Ti) (at.%)
S1	69	16	14
S2	68	18	14
S3	67	19	14
S4	67	26	07
S5	70	20	11
S6	66	25	9
S7	65	30	4
S8	65	30	4
S9	70	26	4

Effects of process parameters on the properties of AlCrTiN coatings

The results of investigations of the hardness and Young modulus, adhesion, and tribological properties of manufactured AlCrTiN coatings are summarized in **Table 5.**

Table 5. Mechanical and tribological properties of AlCrTiN coatings
Tabela 5. Właściwości mechaniczne i tribologiczne powłok AlCrTiN

Specimen no.	Hardness H (GPa)	Young modulus E (GPa)	Adhesion strength L_{C2} (N)	Volumetric wear W_v (mm^3)	Friction coefficient μ
S1	25.8±1.8	298±43	42±4	0.072±0.001	0.65
S2	24.2±1.1	287±14	44±2	0.022±0.005	0.65
S3	25.9±2.1	335±38	43±3	0.033±0.001	0.60
S4	22.9±1.1	283±13	40±2	0.050±0.001	0.65
S5	19.9±2.4	241±25	40±2	0.061±0.009	0.60
S6	21.2±1.1	259±19	35±2	0.051±0.002	0.60
S7	20.7±0.7	291±18	45±1	0.042±0.003	0.65
S8	18.9±1.5	257±21	27±2	0.049±0.002	0.55
S9	18.3±3.4	292±73	24±4	0.070±0.001	0.55

According to the Taguchi method, the effect of the deposition process parameters on the coatings properties were evaluated by analysis of signal-to-noise ratio (SNR) for the each level of selected deposition parameters **(Figs. 2–5)**.

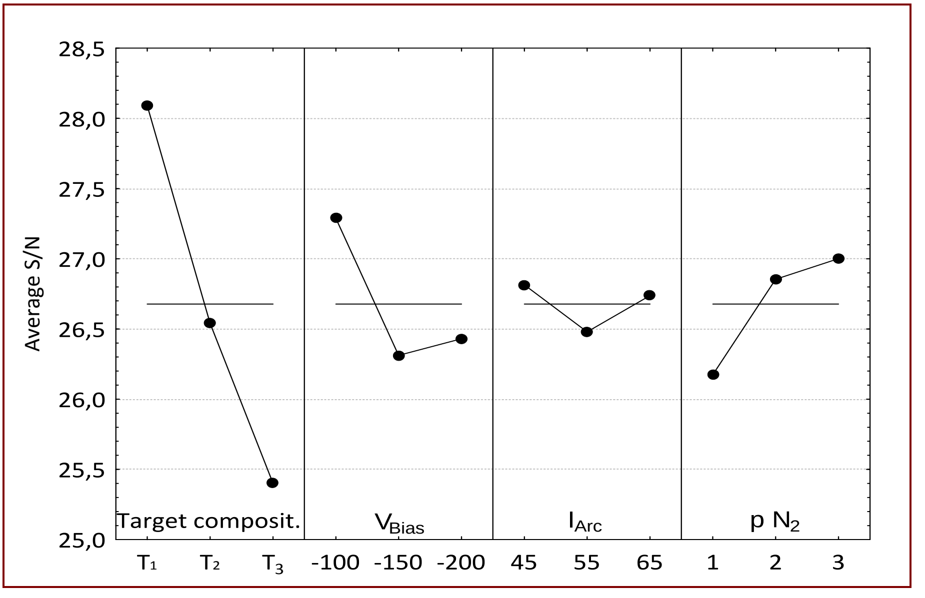


Fig. 2. The effect of each deposition parameter on the hardness of AlCrTiN coatings
Rys. 2. Wpływ poszczególnych parametrów osadzania na twardość powłok AlCrTiN

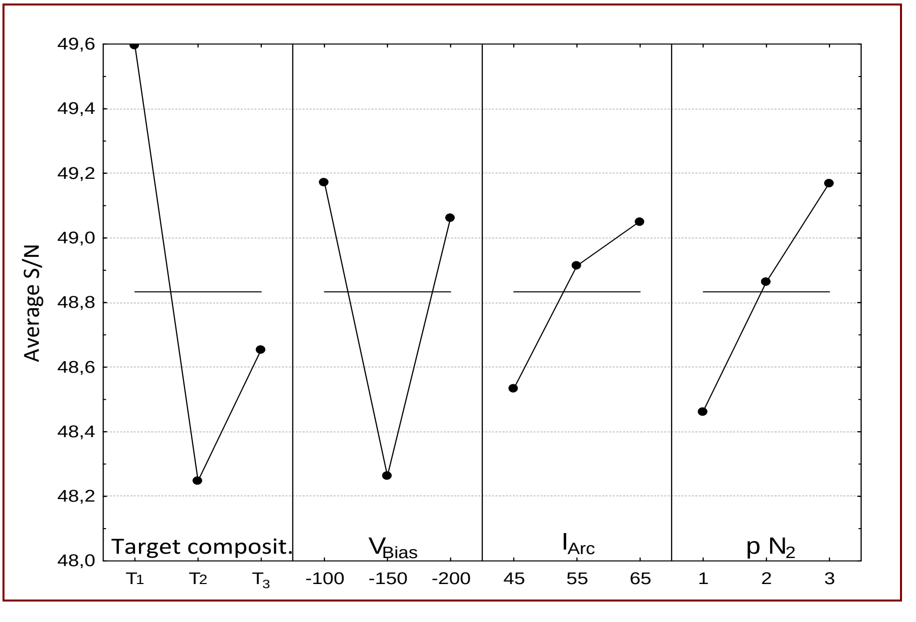


Fig. 3. The effect of each deposition parameter on the Young modulus of AlCrTiN coatings
Rys. 3. Wpływ poszczególnych parametrów osadzania na moduł Younga powłok AlCrTiN

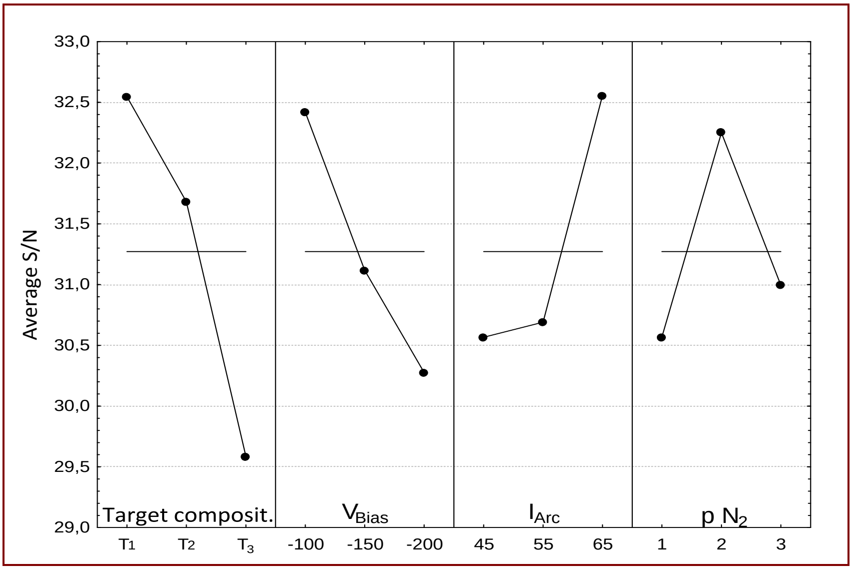


Fig. 4. The effect of each deposition parameter on the adhesion of AlCrTiN coatings

Rys. 4. Wpływ poszczególnych parametrów osadzania na adhezję powłok AlCrTiN

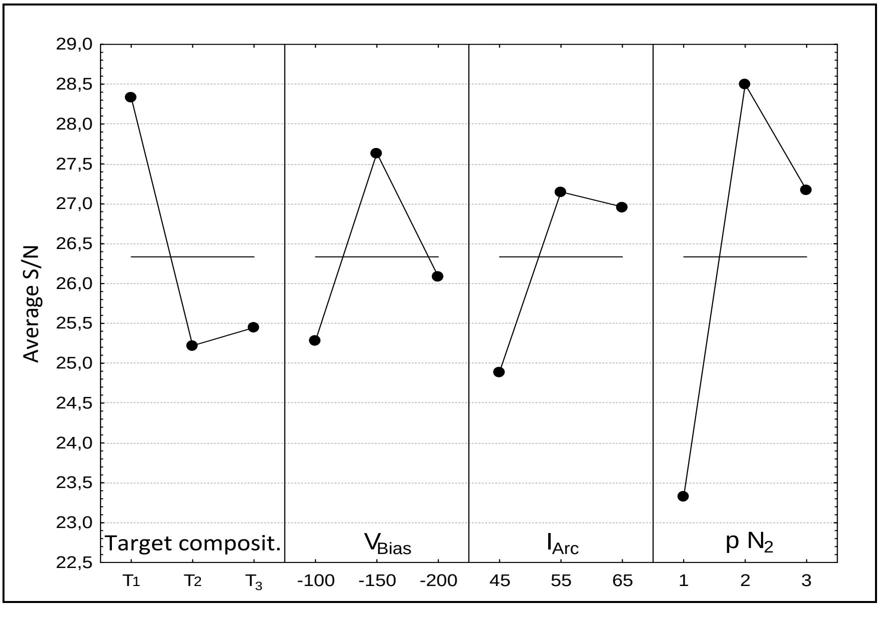


Fig. 5. The effect of each deposition parameter on the volumetric wear of AlCrTiN coatings

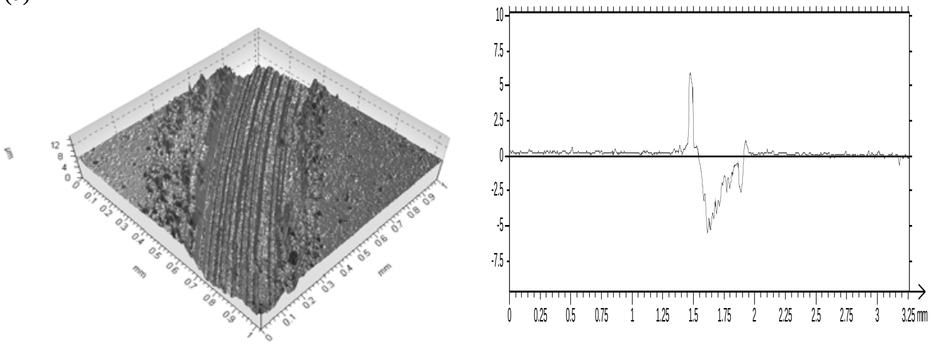
Rys. 5. Wpływ poszczególnych parametrów osadzania na objętościowe zużycie powłok AlCr-TiN

Based on the statistical analyses, the most effective parameters and the optimum deposition condition allowing the production of the AlCrTiN coatings with good mechanical properties and the best wear resistance were determined. In this study, the following optimization criterions were used: „the larger-the-better" (for the evaluation of hardness, Young modulus, and adhesion) and „the smaller-the-better" (for optimising the coatings wear resistance). The maximum signal-to-noise ratio (SNR) in each graph shows the optimal value of the particular parameter to achieve the desired properties of the coating. Based on the analysis of changes the value of signal-to-noise ratio, it was found that, among the tested parameters of the deposition process, the most significant impact on the properties of the AlCrTiN coatings have the following parameters: the chemical composition of targets, the bias voltage, and the pressure of nitrogen. The statistical analysis revealed that the their impacts on the properties of the investigated coatings were as follows: for hardness – 60%, 24%, and 11%, for Young modulus – 46%, 21%, and 20%, for adhesion strength – 27%, 24% and 21%, and for volumetric wear – 30%, 21%, and 34%, respectively.

Analysis of wear mechanism of the AlCrTiN coatings and determination of the optimal conditions of the deposition process in order to improve their wear resistance at elevated temperatures

The results of the tribological tests executed by the ball-on-disc method indicate that, for all the investigated AlCrTiN coatings, the friction coefficient (COF) in the steady-state phase of the run remains practically at a constant level (0.60±0.05). The wear rate of the samples at a temperature of 500°C varies depending on deposition process parameters, but the wear mechanism is the same for all investigated coatings. Many scratch grooves parallel to the sliding direction were observed on the surface of all of the wear track, but their number and depth varied depending on the mechanical properties of the coating **(Fig. 6)**. This mode of abrasive wear (micro scratching) is a generated action of hard particles detached from the coating through the mechanism of crack propagation. Microscopic examinations of the wear tracks showed that that the first micro-cracks appear near the defects existing in the coatings, for example, the microdroplets. The presented type of wear mechanism indicates that the coating with increased resistance to wear should be characterized by both high hardness and good adhesion to the substrate but also and primarily by high fracture toughness. This was confirmed by research results that showed that the minimum values of wear volume W_v = 0.022 – 0.033 mm^3 were recorded on the specimens coated by AlCrTiN coatings with good adhesion to the steel substrates (critical load L_{C2} = 43–44 N), and simultaneously high hardness (H = 24–25 GPa), as well as with a low defectiveness degree of the surface (Sa = 0.07–0.08 μm) (**Table 3, 5**).

(a)



(b)

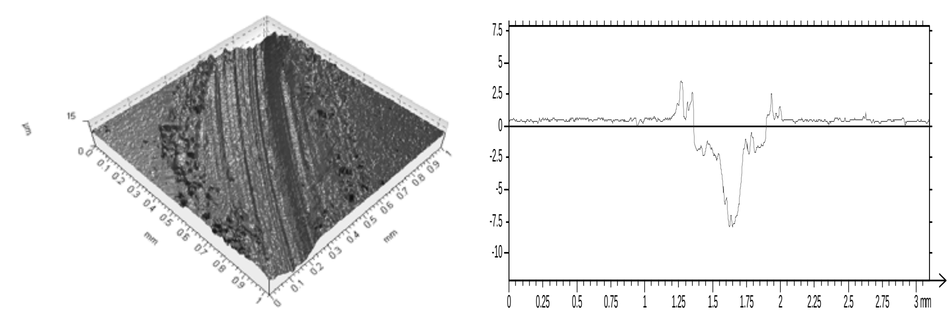


Fig. 6. Topography of the worn surface and the cross-section profile of the wear track on: a) S2 specimen (a lowest wear volume – 0.022 mm^3), b) S9 specimen (a higher volume of wear – 0.070 mm^3)

Rys. 6. Topografia zużytych powierzchni i profil przekroju poprzecznego toru zużycia na: a) próbce S2 (najmniejsza ilość zużycia – 0,022 mm^3), b) próbce S9 (większa objętość zużycia – 0,070 mm^3)

Based on the analysis of the research results performed for the AlCrTiN coatings, it was found that, to obtain high wear resistance at elevated temperatures in combination with the optimal mechanical properties, the cathodic-arc deposition process should be carried out using following parameters: targets with the composition $Al_{0.7}Cr_{0.15}Ti_{0.15}$, a bias voltage of 150 V, an arc current of 55 A, and a nitrogen pressure of 2.0 Pa (Taguchi parameters $T_1A_2B_2C_2$ – **Table 2**).

CONCLUSIONS

The Taguchi method has been applied to study the effect of the selected deposition parameters on the mechanical and tribological properties of AlCrTiN coatings deposited by the cathodic-arc method. Based on the statistical analysis, the most effective deposition parameters that strongly influenced on mechanical and tribological properties of the coatings (i.e. the hardness, Young modulus, adhesion, wear resistance) and the optimum process conditions for the manufacturing of AlCrTiN coatings with elevated properties were determined.

The following conclusions can be drawn based on the study:

- The most effective deposition process parameters to obtain very good mechanical and tribological properties of AlCrTiN coatings are the target composition, substrate bias voltage, and nitrogen pressure.
- To obtain the best high-temperature wear resistance, the coating must have sufficiently high hardness, very good adhesion to the substrate, and low defectiveness of the surface.
- To achieve the best tribological behaviour of AlCrTiN coating, the optimum deposition conditions were the following: the target composition was $Al_{0.7}Cr_{0.15}Ti_{0.15}$, the bias voltage was 150 V, the arc current was 55 A, and the nitrogen pressure was 2.0 Pa.
- The results of the research show that the Taguchi method makes it possible to optimize the manufacturing process to produce tribological coatings with enhanced properties, and it can be used to analyse the influence of the process parameters on the individual properties of these coatings.

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Streszczenie

W artykule przedstawiono wyniki badań nad opracowaniem optymalnych parametrów procesu wytwarzania powłok AlCrTiN metodą łukowo-próżniową, o podwyższonych właściwościach mechanicznych i wysokiej odporności na zużycie ścierne. Na podstawie analizy literaturowej wybrano cztery parametry procesu osadzania istotnie wpływające na właściwości mechaniczne i tribologiczne powłok, a mianowicie: skład chemiczny targetów, ciśnienie azotu, prąd łuku i napięcie polaryzacji podłoży z trzema poziomami wartości. Do badania oddziaływania parametrów procesu łukowo-próżniowego na właściwości powłok oraz przeprowadzenia optymalizacji ich wartości dla uzyskania wzrostu odporności powłok na zużycie, wykorzystano metodę Taguchi'ego. Przy pomocy metod analizy statystycznej określono najbardziej efektywne parametry procesu i optymalne warunki wytwarzania powłok o najwyższej odporności na zużycie ścierne. Wyniki wskazały, że spośród badanych parametrów procesu osadzania istotny wpływ na właściwości mechaniczne i tribologiczne powłok AlCrTiN przede

wszystkim wywierają: skład chemiczny targetów, napięcie polaryzacji oraz ciśnienia azotu. Stwierdzono również, że aby osiągnąć zarówno dobre właściwości mechaniczne, jak i najlepszą odporność na ścieranie powłoki AlCrTiN, proces jej osadzania, z zastosowaniem techniki katodowego niskociśnieniowego wyładowania łukowego, powinien być przeprowadzany w następujący sposób: przy wykorzystaniu targetów Al70% Cr15% Ti15% i zastosowaniu ujemnego napięcia polaryzacji 150 V, prądu łuku o natężeniu 55 oraz ciśnienia azotu 2,0 Pa.