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INFLUENCE OF THERMAL TREATMENT ON THE MECHANICAL PROPERTIES OF TiAlSiN-BASED COATINGS

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ABSTRACT

This work presents a study on the influence of the deposition and annealing temperatures on the mechanical properties of quaternary TiAlSiN-based thin films. The coatings are deposited by cathodic arc evaporation onto substrates of cemented carbide at deposition temperatures of 470°C and 500°C. A part of them is post-deposition annealed at 550°C and 525°C.

The investigation of the mechanical propertied showed strong dependence on the thermal treatment. Highest nanohardness of 45 GPa was obtained at deposition temperature of 500°C and post-annealing at 525°C. The results from the scratch test revealed very good adhesion to the substrates as no delamination was observed, and a relatively low friction coefficient of 0.05-0.07 was obtained.

Keywords: *TiAlSiN coatings; cathodic arc deposition; nanoindentation; adhesion test.*

INTRODUCTION

The nanocomposite coatings are a class of nanostructured materials, which have attracted great interest due to the unique combination of mechanical properties, thermal stability and oxidation resistance. These properties make the nanocomposite coatings a better alternative to the traditional polycrystalline materials, which have met their limits in many recent applications. Currently, the nanocomposite coatings with high hardness, wear- and oxidation resistance, and thermal stability have found wide popularity as protective coatings for many industrial tools [1, 2]. Applied on machining, stamping and forming tools they significantly increase the lifetime and the cutting speed, and consequently - the machining operation productivity [3].

The most important feature of the nanocomposite materials is the hardness exceeding 40 GPa, which is combined with high toughness and adhesion. These properties are important in machining operations on hard and tough materials, especially in extreme industrial conditions like high-speed and dry cutting. Several research groups have focused their investigations on quaternary materials systems, like Ti–Al–Si–N, expecting improvement of the mechanical and oxidation properties in comparison to the usual Ti–Al–N coatings [4]. The TiAlSiN-based nanocomposites consist of nanocrystaline grains with dimensions of 3 - 10 nm, embedded in an amorphous Si_3N_4 matrix [5, 6]. The deposition conditions strongly affect the formation process of nanosized grains and the segregation of amorphous SiN_x phases. Therefore, the study of the mechanical and structural properties of the TiAlSiN-based films on deposition conditions is of importance [7].

The present study is focused on the investigation of the nanohardness and the adhesion of TiAlSiN-based coatings and their thermal treatment during deposition and post annealing.

EXPERIMENTAL

The investigated coatings were deposited on substrates of cemented carbide (WC-Co). Prior to coating deposition, the WC-Co substrates were ultrasonically cleaned in an alkaline solution, rinsed in deionised water and dried at temperature of 130°C.

The coatings are obtained by Cathodic Arc Deposition (CAD) in Platit π80+ equipment. It has innovative Lateral Arc Rotating Cathodes (LARC®) system, which ensures minimization of microparticle generation during evaporation, and consequently - significant improvement of the coating structure and quality. A three axis planetary system provides uniformity of the coating thickness.

Titanium and Al + (18 mass %)Si alloy cathodes are used for deposition of TiAlSiN-based coatings in a nitrogen environment at pressure of $9.0x10^{-3}$ mbar to $4.0x10^{-2}$ mbar. The base pressure is lower than $5x10^{-6}$ mbar. The applied bias voltage is 45 V.

The scheme of the deposited coatings is shown in Fig. 1. In order to improve the coating adhesion, firstly a 100 nm Ti contact layer was deposited.

After that, a TiN gradient contact layer with thickness of 140 nm was formed realizing smooth transition to the TiAlSiN-based coating. The total coating thickness is 3 μm. The TiAlSiN films were prepared by simultaneous evaporation of both cathodes.

Three sets of samples were prepared for characterization of the mechanical properties. The first one denoted #11017-1 was deposited at 470°C. The second set (#11017-4) was deposited again at 470°C, but after that, the samples were annealed for two hours in N_2. The third set, consisted of a test sample (#13029-2) and an industrial tool (#13029), was deposited at 500°C and annealed for two hours in N_2 at 525°C.

The total coating thickness was measured using

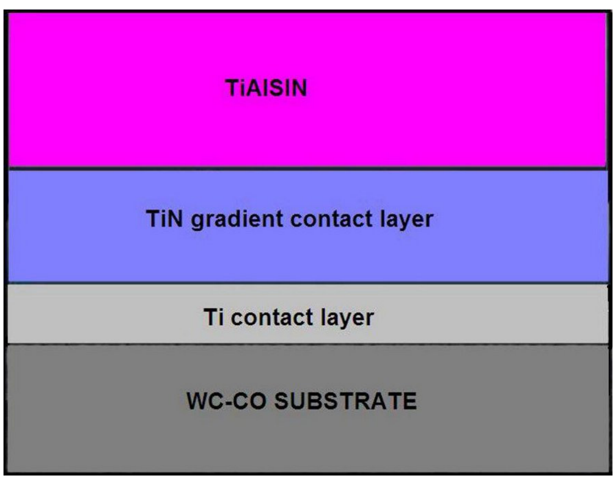

Fig. 1. A principal scheme of the TiAlSiN- based coatings.

a LEITZ Incident-light Microscope Interferometer in monochromatic light (λ=560 μm). The mechanical properties were investigated using a Compact Platform CPX (MHT/NHT) CSM Instruments equipment. The nanohardness and elastic modulus were studied by the Oliver&Phar method. Nanoindentation was performed by triangular diamond Berkovich pyramid in the loading interval of 30 - 500 mN. The scratch tests for coating adhesion and friction coefficient determination were made using a spherical Rockwell indenter with radius of 200 μm and a cone angle of 120° at normal force, progressively increasing from 1 N to 30 N.

RESULTS AND DISCUSSION

Three nanoindentations at each load in the defined interval were done in order to achieve more accurate information. The nanohardness (H) and elastic modulus (E) were determined from the load-depth curves, measured at each loading.

The dependence of the nanohardness on the penetration depth and the applied load is presented in Fig. 2. As it is seen, the coating deposited at 470°C has the lowest nanohardness of 30 GPa. This value is usual for the coatings formed by a solid solution of the reagents. The obtained nanohardness of 30 GPa could be due to the columnar structure of the main coating layer. [8]. Our TEM observations (not presented here) confirmed the presence of a columnar structure, which allows to conclude that the deposition temperature of 470 °C supports the reaction between the cathode material and nitrogen, but is not enough for nanocrystal formation. The samples deposited at the same temperature, but annealed at 550°C for 2 h in N_2, showed somewhat higher nanohardness of 33 GPa. It is known that the increased deposition temperature stimulates the nanograin growth [9]. We suppose, that post annealing at 550°C contributes to the nanostructure formation, however, the nanocrystals are rare and most probably in a size, which is outside the interval limiting superhardness (5 – 10 nm). The samples deposited at 500°C and annealed at 525°C for 2 h in N_2 had the highest nanohardness of 42 - 45 GPa. The increased nanohardness could be due to the Hall-Petch effect, according which, the hardness increases with the particle size decrease [10]. The measured grain size for the coatings deposited at 500°C and annealed at 525°C is between 8 and 10 nm [11].

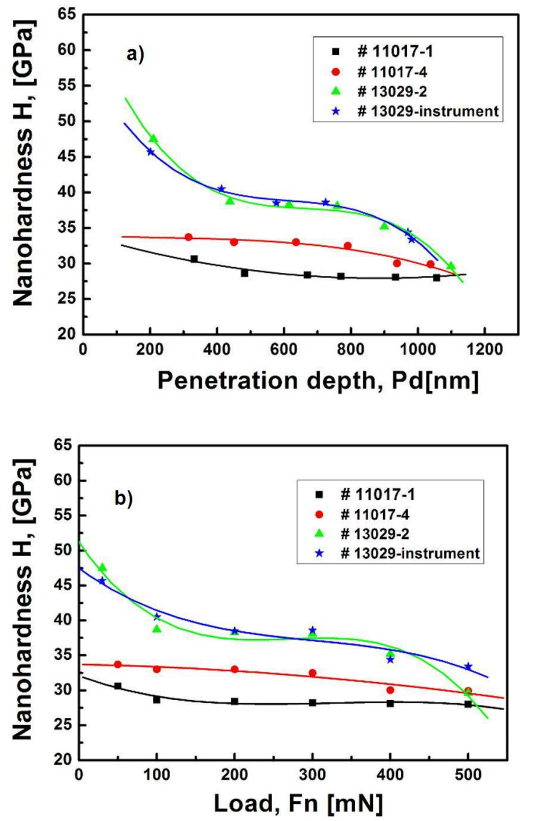

Fig. 2. Dependence of the nanohardness on the penetration depth (a) and the applied load (b) for the investigated samples.

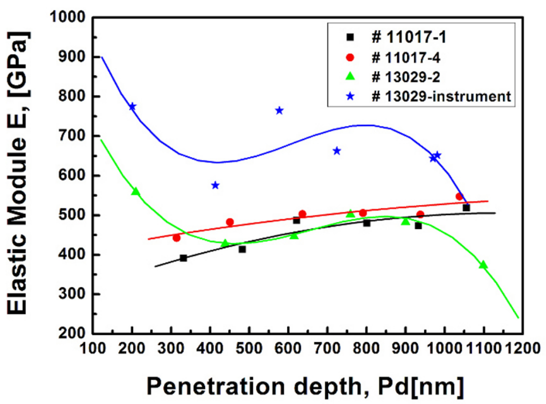

Fig. 3. Dependence of the Elastic modulus on the penetration depth for the investigated samples.

All dependences show decrease of the nanohardness after certain depth, indicating the increased influence of the substrate, which is more pronounced in nanocomposite coatings. The depth, at which the substrate effect appears, is related to the coating hardness.

The dependence of the elastic modulus on the penetration depth is shown in Fig. 3. The measured values are in the range from 300 to 550 GPa. With increase of the penetration depth, the coatings deposited at 470°C show small smooth increase of the elastic modulus value, while for the coatings deposited at 500°C there is a more pronounced change. The XRD and TEM investigations of the samples deposited at 500°C revealed that the silicon nitride in the TiAlSiN coating structure is presented as an amorphous phase [11]. The presence of an amorphous phase suppresses the plastic deformations [12] and leads to higher elastic modulus at the substrate - independent depth.

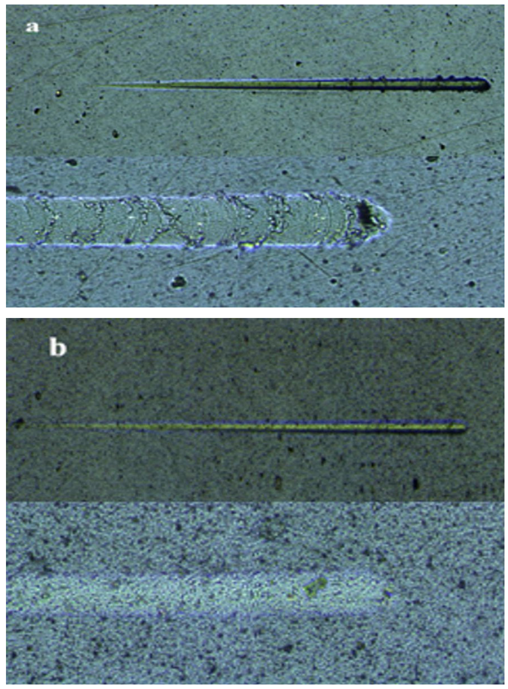

Fig. 4. Scratch tracks pictures observed on top of the TiAlSiN-based samples a) #11017-1 deposited at 470°C, b) #13029-2 deposited at 500°C.

The adhesion and friction coefficient were studied using a micro-scratch test technique. The tests were carried out for the loading force progressively increasing from 1 N to 30 N with a speed of 5 N/s along the path of 3 mm. The critical load Lc, at which features characterized the adhesion, was determined from the value of the measured acoustic emission and the scratch track observed by an optical video microscope.

In Fig.4 the optical images of the scratch tracks of the samples #11017-1(a) and #13029-2 (b) are shown. The sample #11017-1 has very good adhesion and the first circular cracks (Fig. 4a) were observed at loading of 20 N.

Further increase of the loading up to 30 N causes small increase of the circular cracks density, but no coating delamination is detected.

The sample #13029-2 has excellent adhesion and no cracks are observed at loading from 1 N to 30 N. The results of the scratch tests show that the coatings deposited at 500°C and annealed at 525°C in a N_2 atmosphere have better adhesion than the coatings deposited at 470°C. The measured friction coefficients of the examined coatings are within the range of 0.05 - 0.07.

CONCLUSIONS

Three groups of TiAlSiN-based coatings were prepared at different thermal treatments. Correlation between the mechanical properties and the deposition and annealing temperatures was found. The results revealed that the TiAlSiN-based samples deposited at 500°C and annealed at 525°C in N_2 have highest nanohardness. The post annealing causes improvement of the coating structure stability. The achieved hardness of 42 - 45 GPa determines these coatings as superhard.

The scratch test shows that the nanocomposite coatings have excellent adhesion and even at applied normal force of 30 N, no damages are observed.

The results of the mechanical properties examination show that the thermal treatment of the TiAlSiN – based films is very an important technology parameter to achieve coatings appropriate for applications in high speed machining.

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