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CHANGING THE INTEGRITY OF THE MATERIAL SURFACE BY COMBINING LASER SURFACE TEXTURING AND PVD MAGNETRON SPUTTERING TECHNOLOGIES

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Abstract. The presented research combines laser micromachining technology (LST, Laser Surface Texturing) using an ultra-short femtosecond laser beam and PVD deposition of thin layers using magnetron sputtering. This combination has multiple benefits. It provides uniform and highly adhesive coatings with improved tribological properties. By optimising the deposition parameters when applying a thin layer of TiN, an improvement in the tribological properties of the tool steel surface and the laser micromachining (LST) surface can be achieved. On the laser-modified and coated surface with Ellipses-texture, the wear reduction was almost 19 % compared to the reference texture. The TiN coating exhibited very good adhesion to the substrate with critical force values of L_{C3} greater than 45 N and a nano hardness value of 26.5 GPa. The adhesion of the TiN coating to the laser-affected substrate, as well as the resistance of the coating to damage, was high. Damage primarily occurred on the original polished surface around the edges of the machined textures, where stresses are likely to concentrate.

Keywords: PVD magnetron sputtering, laser surface texturing, mechanical and tribological properties.

1. Introduction

Surface integrity refers to a surface's inherent or enhanced condition resulting from machining or other surface-generating operations. Wear, abrasion, and tribology behaviour are common issues associated with this [1]. Changing the surface topography can effectively improve tribological behaviour. In dry sliding conditions, the primary effect is removing or trapping wear debris or particles from the contact. When lubrication is present, the micro-texture can function as a reservoir for liquid lubricants, effectively delivering the lubricant between the sliding surfaces and causing positive changes in friction and wear [2, 3].

Using LST (laser micromachining), surface topography or microstructures on large parts can be prepared. Laser surface texturing (LST) replaces traditional techniques like surface extrusion or blasting. Laser systems can be tailored to accommodate various materials and specific application needs. LST presents a promising process due to its rapid speed, eco-friendly nature, and ability to precisely manipulate the shape and size of micro-dimples [4]. Femtosecond lasers are now widely utilised for their precise ability to modify material surfaces with minimal thermal impact. The textured surface of tools created with these lasers showed improved coating retention during turning experiments, ultimately extending the lifespan. However, the substrate also plays a significant role [5].

Applying PVD coatings to the surface of conventional machining tools enhances their wear resistance, leading to improved cutting efficiency and higher processing quality. Due to the different ways the coatings bond with the substrate, their thermal expansion coefficients, and their wetting properties, PVD-coated tools often experience problems with insufficient adhesion of the coating. To fully benefit from PVD coatings, ensuring strong adhesion between the coatings and the substrate is important. The adhesive strength between the coatings and the substrate is influenced by factors such as the chemical bonding properties at the coatings-substrate interfaces, specific surface area, topography, and substrate surface energy [6, 7]. In general, several methods have been used to enhance adhesion at the substrate-coating interface. These methods include substrate pre-treatment, intermediate layers, multilayer coatings, and functional layers [7]. However, applying the adhesive interlayer necessitates strict equipment conditions [6]. Making the surface rougher can help improve the adhesion of PVD coatings because it enhances the mechanical interlocking between the coatings and the substrate [4].

In the area of PVD tool coatings, there is a rising interest in enhancing coating adhesion through LST of the substrate surface. This process aims to increase the contact area between coatings and the substrate, thereby improving mechanical anchoring at the interface [6]. Studies have demonstrated that laser treatment is more effective than abrasive flow machining for enhancing coating adhesion [8]. Also, in [7], they discovered that LST led to enhanced adhesion of PVD coatings compared to the traditional cleaning method of grit blasting.

Hard coatings like TiN, TiCN, and TiAlN are commonly used in the metal-mechanics industry to enhance the properties of machine parts and cutting tools. TiN is widely used as a coating due to its high hardness and chemical stability. Ti-based coatings have been shown to reduce the wear of basic material by approximately four times [1]. The effect of using laser processing to create surface textures on substrates and how it impacts the interfacial properties of PVD tool coatings, particularly the physical-chemical bonding interfaces, still needs to be clarified.

This work aims to explore the possibility of improving tribological properties in dry sliding conditions (economically and environmentally advantageous) by combining surface LST texturing and hard TiN coating. Magnetron sputtering is an effective method for enhancing material surface properties by creating uniform thin layers that adhere well to the base material. The combination of laser structuring and magnetron sputtering is expected to enhance the material's useful properties while preserving its integrity.

2. Materials and methods

The substrate samples were made of tool steel EN ISO HS6-5-2C. Tool steel discs with diameters of ⌀20 mm × 5 mm were used as a substrate. Their nominal composition is as follows (in wt.%): 0.9 C; 0.31 Mn; 0.34 Si; 0.026 P; 0.0005 S; 4.43 Cr; 4.78 Mo; 5.93 W; 1.79 V; and 0.65 Co. The discs were hardened to 64-65 HRC and polished to a roughness (Sa) of 0.01 µm.

The samples' surfaces were modified using short-pulse femtosecond lasers through the Laser Surface Texturing (LST) method at the CIIRC at CTU. Textures of various shapes, including pits (Pit), hexagons (Hex), ellipses with a depression (Eli–), ellipses with a protrusion (Eli+), and pyramids (Pyr), were produced on the tool steel samples. The depth of the texture is approximately 10 µm for all samples.

A thin layer of TiN was deposited on the laser-modified surfaces using magnetron reactive sputtering technology. Two different recipes (B21 and B22) were utilised to prepare the thin coating to achieve optimal deposition parameters of the coating (see below).

The coating thickness (D) is determined using a Calotest method, which involves a 30 mm steel ball and 0.1 mm monocrystalline diamond grains in diamond paste. The average value of the thickness of the thin films was calculated from three measurements on the surface of each coating.

The local chemical analysis of the coatings was carried out using a Zeiss Ultra Plus scanning electron microscope equipped with an Oxford X-Max 20 energy dispersive spectrometer. The analyses were conducted at an accelerating voltage of 10 kV and a current of 1.6 nA, with a working distance from the sample surface of 8 mm and pulse pile-up correction.

The surface morphology was estimated using a SENSOFAR S Neox confocal microscope. The scanned surface area of the samples was 850 × 709 µm. The average value of the surface roughness of the thin films was calculated from the five measurement on the surface of each coating.

Nanohardness is measured using a Berkovich diamond indenter prism on CSM Instruments equipment. The maximum penetration depth is limited to 10 % of the coating thickness. The values of nanohardness and elastic modulus of the thin layers were obtained from nanoindentation, with 25 indentations performed on each sample. Subsequently, the plasticity index (H/E) and the material's resistance to plastic deformation (H^3/E^2) are calculated from the measured values.

The adhesion and toughness of the coatings are evaluated using a micro-scratch test technique, using a Rockwell diamond indenter with a 200 µm tip radius to create scratch adhesion tracks. The evaluation involved gradually increasing the tip load from 2 to 60 N while simultaneously moving the sample linearly in a perpendicular plane to the indenter. The tracks are then analysed using an optical microscope. Three measurements were taken at different places, and the average values of the measured critical loads (L_C, Load Critical) were calculated. The major failure events have been classified in terms of plastic deformation, cracking (L_{C1}), spallation (where the coating flakes off, typically at the edges) (L_{C2}), and penetration of the coating to the substrate at the track centre (L_{C3}), see EN 1071-3:2005 [9]. The critical load L_{C1} is the point on the graph where the coating's first crack (cohesive failure) is observed. L_{C3} is the point on the graph where the damage becomes continuous, leading to complete coating delamination. This is accompanied by a sharp increase in the coefficient of friction (CoF). L_{C2} represents the force at which 50 % of the coating is damaged; L_{C2} has not been evaluated. The average value of thin film adhesion was calculated from three measurements on the surface of each coating.

The tribological measurements were conducted using a TRB^3 tribometer from Anton Paar under dry sliding conditions. The measurements were taken at room temperature (RT) following the ASTM G99 standard. The parameters of the tribological experiment were as follows: Ball-on-Disc method, 10 N load, counter-body diameter of 3 mm, track radius on the disc of 3 mm, 120 rpm, and a total track length of 100 m. The counter-body used was a ceramic ball made of Al_2O_3, with a hardness of 2100 HV and a density of 3 860 g cm^{-3}. Tribological measurement was performed once on the surface of each sample. The surface wear of the friction pairs was assessed using a confocal microscope to measure the depth and width

Recipe	D [μm]	d [nm]	H [GPa]	H [HV]	E^* [GPa]	H/E [-]	H^3/E^2
REF19 + B21	0.8	81	30.2 ± 5.4	2 794 ± 501	401.2 ± 23.2	0.075	0.171
REF19 + B22	0.9	93	26.5 ± 2.3	2 458 ± 211	379.0 ± 22.9	0.070	0.130

TABLE 1. Mechanical properties of TiN thin layers.

of the wear of the tribological track at a distance of 90° axially.

3. Results and discussion

3.1. Magnetron deposition of thin layer

A thin layer of titanium nitride (TiN) was selected to modify the surface of the tool steels. Initially, the deposition parameters of the magnetron reactive sputtering process needed optimisation. The chemical composition of the coating during deposition is mainly influenced by the parameters of the working pressure, flow rates, gas ratios, and the pulse source's power. Parameters of deposition of TiN thin layers: after reaching a primary vacuum of 8×10^{-4} Pa, the substrate was pre-cleaned in an argon plasma at a working pressure of 3 Pa or Ar gas flow 6.5 sccm and power 1 kW for 40 min. Then an adhesive intermediate layer of pure titanium was sputtered at a working pressure of 0.3 Pa or with an Ar flow of 46 sccm and a source power of 1.5 kW for 5 min. A circular target with a diameter of 77 mm with a Ti purity of 99.99 % was placed on the cathode. Reactive sputtering of TiN was carried out with an Ar:N flow rate of 8:1 sccm at a voltage range of 1000 V for B21 and 1500 V for B22 for 1 and 3 h.

The thickness of the thin coating can affect the magnitude of the residual stress generated in the structure. Therefore, the reaction time of the TiN process has been optimised to a range of one, two, and three hours. The measured coating thicknesses for the B21 recipe are as follows: 2.6 μm, 1.7 μm, and 0.8 μm for a 3-hour, 2-hour and 1-hour deposition, respectively. For the B22 recipe, the coating thicknesses are: 3.5 μm, 1.8 μm, and 0.9 μm for 3-hour, 2-hour, and 1-hour depositions, respectively.

The optimal deposition process for the TiN coating on LST was chosen for deposition times of 1 (labelled B21/1 and B22/1) and 3 hours (labelled B21/3 and B22/3) using formulations B21 and B22. The recipes B21 and B22 used for depositing the TiN coating showed, based on EDX chemical composition analysis, a stoichiometry (ratio of Ti and N) in the equilibrium atomic ratio. The criteria for choosing the optimal thin coating application process were achieving good mechanical properties of the coatings, i.e. achieving good adhesion, nano-hardness and reducing layer wear.

Recipe	L_{C1} [N]	L_{C3} [N]
REF19 + B21/1	7.6 ± 0.9	31.2 ± 1.6
REF19 + B21/3	8.5 ± 0.5	40.4 ± 1.9
REF19 + B22	8.2 ± 0.5	49.8 ± 1.7

TABLE 2. Evaluation of adhesion critical forces of TiN thin layers.

Sample Recipe	Disc wear width [μm]	Disc wear depth [μm]	CoF μ [-]
REF19	388.2 ± 4.4	3.7 ± 0.2	0.55 ± 0.06
REF19 + B21/1	119.8 ± 21.4	0.2 ± 0.0	0.23 ± 0.04
REF19 + B21/3	119.2 ± 4.1	0.1 ± 0.0	0.23 ± 0.05
REF19 + B22	112.2 ± 2.7	0.1 ± 0.0	0.22 ± 0.02

TABLE 3. Results of tribological analysis of TiN thin layers.

3.2. Mechanical properties of thin coatings, evaluation and selection of deposition parameters

In the tables, the reference polished tool steel is labelled as REF19. The results of the nanoindentation of thin coatings were statistically processed; the average values with standard deviation are presented in Table 1, denoted as follows: coating thickness (D), indentation depth (d), nanohardness (H), Young's modulus of elasticity (E), plasticity index (H/E), and material resistance to plastic deformation (H^3/E^2).

Table 2 shows the measured values of the critical forces L_{C1} and L_{C3}, providing information about the coatings' adhesion to the substrate as evaluated by the scratch test method. Table 3 shows the results of the tribological properties of the modified surfaces.

3.3. Microscopy and image analysis of laser-textured thin-layer surfaces

Confocal microscopy (Figure 1) was utilised to assess surface morphology and roughness. Image analysis was conducted using Matlab software (The MathWorks, Inc.). An automated program code was employed to determine the machined area, the perimeter of the detected object, as well as the length, width, and aspect ratio of the object (results shown in Table 4).

For the "Pit" pattern, the length is approximately 1.25 times longer than the width, with a machined area of up to 20 %. For Eli–, the length is approximately

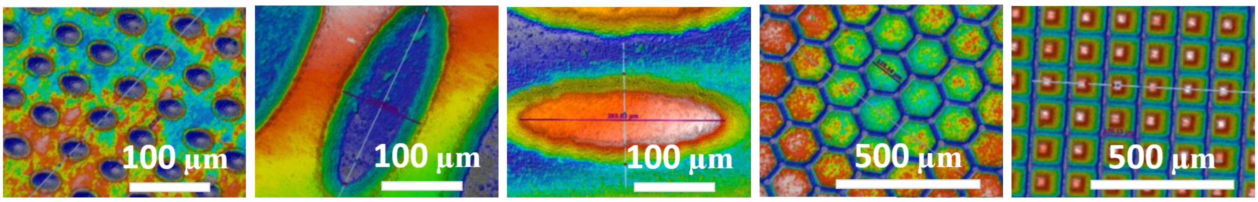
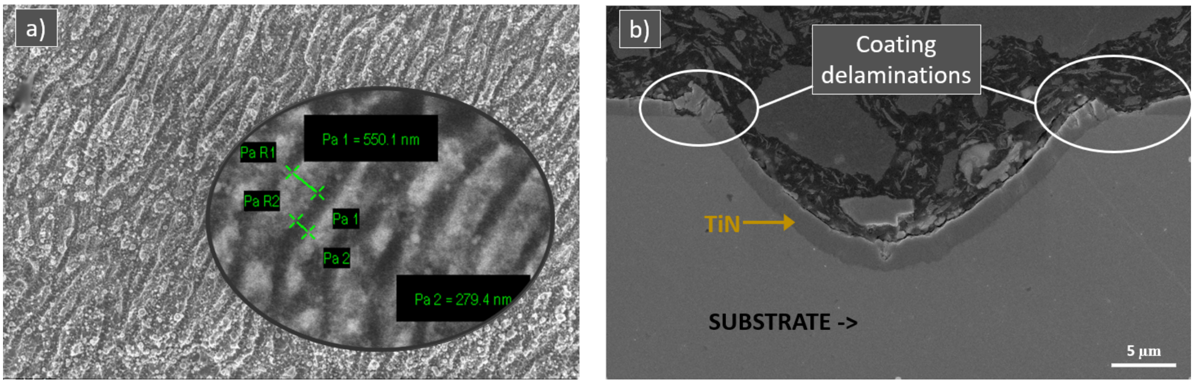


FIGURE 1. Confocal microscopy of the surface REF19 samples with surface texture type: Pits, Ellipse-, Ellipse+, Hexagon, Pyramids. Images are depicted on a height scale, with blue representing valleys and red colour representing peaks.

LST structure	LST area [%]	LST area [µm^2]	Perimeter [µm]	Length [µm]	Width [µm]	Aspect ratio [–]
Pit	19.9 ± 1.1	735.5 ± 41.9	102 ± 3.8	34.2 ± 1.5	27.2 ± 1.3	1.26 ± 0.07
Eli–	44.5 ± 1.0	32 082 ± 1 028	791 ± 8.8	331 ± 2.9	123.5 ± 3.2	2.68 ± 0.06
Eli+	69.8 ± 4.2	21 099 ± 389	669 ± 6.2	286 ± 3.7	93.4 ± 1.3	3.09 ± 0.06
Hex	43.6 ± 10.3	9 745 ± 2 917	369 ± 95.4	107 ± 26.9	99.3 ± 25.4	1.12 ± 0.11
Pyr	71.9 ± 10.0	3 995 ± 646	243 ± 28.4	73.7 ± 8.1	64.5 ± 6.9	1.18 ± 0.10

TABLE 4. Results of evaluation of object parameters in the image using image analysis.



(A). (B).

FIGURE 2. SEM image of a) ablation trace "LIPSS- Laser Induced Periodic Surface Structure" (mag. 7 000X) and b) metallographic cut of Pit texture with B21/3 coating (mag. 2 500X).

2.7 times longer than the width, with a machined area of up to 45 %. For Eli+, the length of the texture is approximately 3 times longer than the width, with a machined area of up to 70 %. The Hex and Pyr textures are almost symmetrical, with an aspect ratio parameter close to 1 and a machined area of approximately 44 % and 72 %, respectively.

An SEM was used to capture images to analyse the morphology of the ablation trace and thin layer (Figure 2). The EDS method was utilised to determine the chemical composition of the thin layers. To observe the uniformity of the thin layers in conjunction with LST modification, metallographic slices were prepared by cutting the sample.

Even though the results on the surface of the tool steel with the B21 (REF19 + B21) recipe showed favourable tribological properties (Table 3), the coatings on LST samples exhibited delamination (Figure 2b), possibly caused by high residual stresses. As a result of this issue, the TiN coating deposition recipes were further optimised.

3.4. Tribological properties of thin layers deposited on Laser Surface Texturing surfaces

The tribological analysis of thin layers deposited on Laser Surface Texturing surfaces revealed a low coefficient of friction compared to reference sample REF19 (Table 5). At the same time, an increase in the wear depth of the material was observed. Of all the textures examined, only the Pit texture showed lower wear than REF19, even though there was an increase in CoF (Table 5). In an experiment that combined LST and PVD technologies, all LST samples were coated with a thin layer of TiN following the B21/3 recipe. While the tribology results for the tool steel surface REF19 + B21 recipe showed favourable tribological properties (refer to Table 3), the coatings on the LST specimens exhibited delamination, likely due to high residual stress.

Confocal and electron microscope images confirm the occurrence of delamination along the edges of the LST texture. High residual stress along these edges is

Sample	**Disc wear**		**CoF**
LST structure	**width** [μm]	**depth** [μm]	μ [-]
REF19	388.2 ± 4.4	3.7 ± 0.2	0.55 ± 0.06
Pit	319.5 ± 4.9	2.5 ± 0.1	0.62 ± 0.03
Eli–	382.0 ± 5.4	3.9 ± 0.2	0.48 ± 0.03
Eli+	491.1 ± 31.3	12.2 ± 1.0	0.46 ± 0.03
Hex	362.1 ± 5.9	4.3 ± 0.1	0.47 ± 0.02
Pyr	448.4 ± 26.3	9.3 ± 0.4	0.58 ± 0.03

TABLE 5. Wear and friction coefficient of tool steel with LST modification without coating.

Sample	**Disc wear**		**CoF**
LST structure + Recipe	**width** [μm]	**depth** [μm]	μ [-]
Pit + B21/3	351.2 ± 9.8	4.6 ± 0.1	0.57 ± 0.06
Hex + B21/3	400.9 ± 16.1	5.9 ± 0.1	0.61 ± 0.03
Eli– + B21/3	465.3 ± 9.3	5.0 ± 0.2	0.56 ± 0.04
Pyr + B21/3	528.1 ± 6.8	7.3 ± 0.3	0.57 ± 0.06
Eli+ + B21/3	432.1 ± 21.0	10.7 ± 0.4	0.55 ± 0.06
Eli+ + B21/1	430.9 ± 23.9	9.6 ± 0.4	0.42 ± 0.04
Eli– + B22/1	321.8 ± 48.1	3.2 ± 0.3	0.46 ± 0.04

TABLE 6. Wear and friction coefficients of LST samples coated with PVD thin layers.

the likely cause of the delamination. (Figure 2b and similarly in [10]). There was a need to further optimise the recipes used for applying the TiN coating in two ways: a) by reducing the coating thickness (REF19 + B21) and b) by using different recipes (REF19 + B22). Table 6 shows the tribological behaviour of the LST specimens combined with the thin layers. They show a reduction in wear compared to the condition of the base material and the original recipe B21/3.

On the coated and laser-treated surface with Eli texture and thin layer (formulation B22), wear was reduced by almost 19 % compared to Eli texture on the base material. However, damage to the coating was observed during the tribological test. The TiN coating exhibited good adhesion to the substrate, with critical force values of L_{C3} greater than 45 N and a nanohardness value of 26.5 GPa.

The wear reduction on the surface of Eli+ and Pyr samples was achieved despite defects on the surface of the B21/3 recipe. These samples demonstrated a reduction of wear of approximately 12 % and 21 % (on the coating with reduced thickness Eli+ with B21 thin layer, a 21 % reduction in wear was observed) compared to the textures on the base material. Textures Eli+ and Pyr have a more exposed surface and a larger laser-machined area, respectively, compared to the other textures (Table 4). Also, in [5], the LST applied on a larger fraction area of tool steel surface positively affected decreasing the coefficient of friction.

The TiN coating has strong adhesion to the laser-machined surface and is highly resistant to damage. The damage mainly occurs on the original polished surface around the edges of the machined textures, where stresses are likely to be concentrated. The significant increase in surface damage of 27 %, 36 %, and 84 % for the Eli–, Hex, and Pit textured surfaces with the B21/3 coating, compared to the textures on the base material, was likely caused by the combination of higher-thickness coatings (which are brittle and hard, about 30 GPa) with high internal stress.

4. CONCLUSION

By optimising the deposition parameters and applying a thin layer of TiN, a significant improvement in the mechanical and tribological properties of the surface of the tool steel and other surfaces that were processed by laser micromachining technology (LST) was achieved.

In accordance with the B22 recipe, applying a TiN coating on the tool steel increased damage resistance on the polished surface by over 31 times in terms of depth compared to the reference surface. Additionally, the friction coefficient was reduced by more than half (by approximately 60 %).

The wear on the laser-modified surface coated with Eli– texture was reduced by almost 19 % compared to the reference texture despite the coating being damaged during the tribological test. The TiN coating also exhibited good adhesion to the substrate, with L_{C3} critical force values exceeding 45 N and a nanohardness value of 26.5 GPa.

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