Influence of Surface Substrate Texture on the Properties of Al_2O_3 / IF-WS_2 Surface Layers

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Abstract. Anodic oxidation of aluminum alloy in a ternary solution of SAS (sulfuric, adipic and oxalic acids) with inorganic fullerene-like tungsten disulfide (IF-WS_2) is named in the article Al_2O_3/IF-WS_2. The thickness, geometric structure of the surface (SGP) and the tribological properties such as friction coefficient of Al_2O_3/IF-WS_2 junction with polieteroeteroketon filled with graphite, carbon fiber and PTFE (named PEEK/BG) were investigated. The influence of electrolysis time and temperature on the tribological properties of coatings was studied using 2^k factorial design.

Introduction

Surface texture is generally understood to mean the nature of a surface. In the literature, surface structure tends to be used to refer to surface roughness, characteristics of layer and waviness [1]. It is common knowledge that surface topography is one of the most important factors that control friction and transfer layer formation during sliding. For this reason, a great research effort has been directed towards understanding the effect of surface texture on properties of different kind of materials. Due to their properties, aluminum and its alloys have become one of the most commonly used materials in different kinds of industry. Aluminum alloy has many uses for front and functional surfaces, which come into contact with other materials and weather conditions, therefore processes for its surface treatment have become a critical issue in research works. Kadleckova and co-workers [2] reported that with the multistep etching process, the aluminum-alloy substrate can be effectively modified and textured to the same morphology, regardless of the initial surface roughness. In [3] the authors investigated the effects of surface roughness on the tribological properties of a textured surface. Their investigations show that in contrast with dimple textures, surface roughness is a texture at the micro-level, which will essentially influence the load-bearing capacity of lubricant film. In recent years there has been a growing interest in roughness measurement problems which are important for tribological tests [4]. The hard anodizing process of aluminum alloy is a commonly discussed type of surface technology [5-7]. Thin aluminum-oxide films have a great importance for applications in kinematic friction nodes in dry lubrication (i.e. pneumatic cylinders).

Statistical experimental design methods have been shown to be an efficient technique to different coatings property description [8, 9]. One of these methods is a two-level factorial design which involves simultaneous adjustment of experimental factors at high and low levels. It is well known that a full factorial design may also be called a fully crossed design. Such an experiment allows for analyzing the effect of each factor on the response variable, as well as the effects of interactions between factors on the response variable. The most widely-used type of correlation coefficient is Pearson *r*, also called a linear or product-moment correlation and it is allow for the measurement of the degree of relationship between linearly related variables.

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In this paper, the influence of electrolysis time and temperature on the tribological properties of coatings was studied using a 2^k factorial design. The thickness, geometric structure of the surface (SGP) and the tribological properties such as friction coefficient of Al_2O_3/IF-WS_2 junction with polieteroeteroketon filled with graphite, carbon fiber and PTFE (named PEEK/BG) were investigated.

The rationale for this study is the importance attached to the durability of coatings, especially those subjected to tribological loads. It is of interest to many industrial sectors, in particular power hydraulics used in heavy working machines [10-12], including industrial robots [13], as well as heat energy transport [14, 15] and parts working in chemically aggressive biotechnological environments [16, 17]. The methodology used should also be interesting for related surface improvement methods, e.g. electro-spark deposition [18, 19] and laser machining [20]. It can also significantly affect the methods of image analysis [21] and the decision inference schemes [22, 23].

Experimental part

Methodological bases. Macroscopic images of the samples were made with an Olympus BX60M optical microscope with a Motic camera. The thickness of the layers was measured with a Dualscope MP40 by Fischer, using the eddy current method. 10 measurements were performed along the length of the sample and then the average value was calculated. The structural geometry parameters (SGP) measurements of oxide layers were made by the Taylor Hobson Talysurf 3D pin profilometer with the accuracy of 2%. The results of the parameters were developed by means of the TalyMap Universal 3D software. The stereometric analysis was performed on an area of 2 mm x 2 mm. A 2k factorial design with one repetition was applied for analyzing the influence of the surface substrate texture and temperature on thickness, friction coefficient, wear intensity of PEEK/BG plastic and roughness of Al_2O_3/IF-WS_2 coatings. The statistical analysis of the result was performed using the STATISTICA 12.0 software. Tribological measurements were performed on a T17 tester (made by ITE BIP Radom), a pin-on-plate in a reciprocating motion, at room temperature, at the humidity of 30±5%, using 0.5 MPa pressure at an average sliding speed of 0.2 m/s in dry friction conditions. The tribological test was conducted for the sliding distance of 15 km. The commercial PEEK/BG plastic pin with a diameter of 9 mm was used as a counter-body. The friction coefficient was measured when a steady state was reached in the friction test. The wear quantity of the PEEK/BG plastic was studied by means of a WA32 analytical balance with the accuracy of 0.1 mg, after each friction process.

Sample preparation. EN-AW-5251 aluminum alloy was the starting material for the process. The samples were etched sequentially with 5% KOH solution for 45 minutes, and 10% HNO_3 solution for 10 minutes, at room temperature. After each step of etching, a sample was placed in distilled water to remove residual acid. The electro-oxidation of the aluminum alloy was carried out in a SAS ternary solution (18% sulfuric (33 ml/l), adipic (67 g/l) and oxalic acids (30 g/l)) with admixture of 15 g of the commercially available IF-WS_2 nanoparticles (NanoMaterials Ltd) per 1 liter of electrolyte. The hard anodizing process was performed at 3 A/dm^2 current density. In order to ensure homogeneity of the suspension and to prevent the settling of IF-WS_2 nanopowder, mechanical stirring was applied during the electrolysis process. The details concerning the serial number of samples according to 2^k factorial design are presented in Table 1. It was decided that the independent variables for this investigation were two levels of temperature of electrolysis and surface of substrate and dependent variables were the thickness, friction coefficient, Ra surface roughness parameter and wear intensity of PEEK/BG. For the purpose of this article, the type of surface substrate was called "horizontal" for samples whose surface addicted by the production of aluminum alloy was parallel to the direction of motion in the tribological test and called "vertical" for perpendicular direction of motion.

Results and discussion

The results of tests can be seen in Table 1. These tests showed that the oxide thickness takes values from 22.50 μm for sample 2 to 27.45 μm for sample 4 (Table 1).

Table 1. Factor settings and results of test for 2^k factorial design

Factor/ sample name	Independent variables		Dependent variables			
	Temperature (°C)	Surface of substrate	Oxide thickness (μm)	*Ra* before tribological test (μm)	Friction coefficient μ	Wear intensity of PEEK/BG
1	30	horizontal	26.62±0.35	0.33±0.05	0.129±0.001	0.124±0.008
2	25	horizontal	22.50±0.44	0.75±0.13	0.271±0.002	0.279±0.014
3	30	vertical	24.61±0.43	0.19±0.06	0.126±0.002	0.064±0.009
4	25	vertical	27.45±0.33	0.31±0.07	0.273±0.003	0.504±0.008

The scatter chart of oxide thickness and *Ra* roughness parameter over type of structure of substrate and temperature of anodizing process are depicted in Figure 1a and b respectively. In order to measure the statistical relationship between two continuous variables, the Pearson's correlation coefficient *r* was used (Eq.1). Pearson's coefficient gives information about the magnitude of the association, or correlation, as well as the direction of the relationship.

$$r = \frac{n\sum xy - \sum x \sum y}{\sqrt{[n\sum x^2 - (\sum x)^2][n\sum y^2 - (\sum y)^2]}} \qquad \text{(Eq. 1)}$$

where: n – number of variable, x, y- variables.

The correlation coefficient represents the strength of an association and is graded from zero to 1.00. It has no units, but may be positive or negative. The Table 2 provides a rule of thumb scale for evaluating the correlation coefficient. In Table 3, the values of correlation coefficient between the analyzed variables are shown.

Our experiment demonstrated little if any correlation of oxide thickness with the temperature of anodizing process and a low negative correlation with the type of structure surface. Using both temperatures, the oxide with the thickness of above 20 μm could be obtained, which is a satisfactory value for a tribological test. As shown in the results (Table 3) of *r* coefficient for *Ra* roughness parameters before tribological test, one could observe a moderate correlation with temperature and the type of structure substrate. The sample obtained in the lower temperature and with the horizontal structure of substrate exhibited the highest value of *Ra* roughness parameters, while the lowest one value of *Ra* was noticed for the higher temperature and vertical structure of substrate.

The further analysis of tribological properties showed that the higher values of friction coefficient μ between Al_2O_3/IF-WS_2 surface layer and PEEK/BG pin were achieved by samples 2 and 4. The graphs which show dependence of friction coefficient μ versus sliding distance is shown in Figure 2a, the scatter chart of friction coefficient in Figure 2b and wear intensity of

PEEK/BG plastic in Figure 2c. Pearson's correlation coefficient (Table 3) between friction coefficient μ and temperature equals r = - 0.99, which means very a high negative correlation. A significant correlation was revealed also for friction coefficient and wear intensity and it equalled r=0.88. The most surprising correlation was r = -0.003, which means that there is any correlation between friction coefficient μ and the type of structure of a substrate. There was no correlation between friction coefficient μ and thickness of oxide, either. The friction coefficient correlates with moderate strength with *Ra* roughness parameter, therefore it can be assumed that there is certain dependence between those variables.

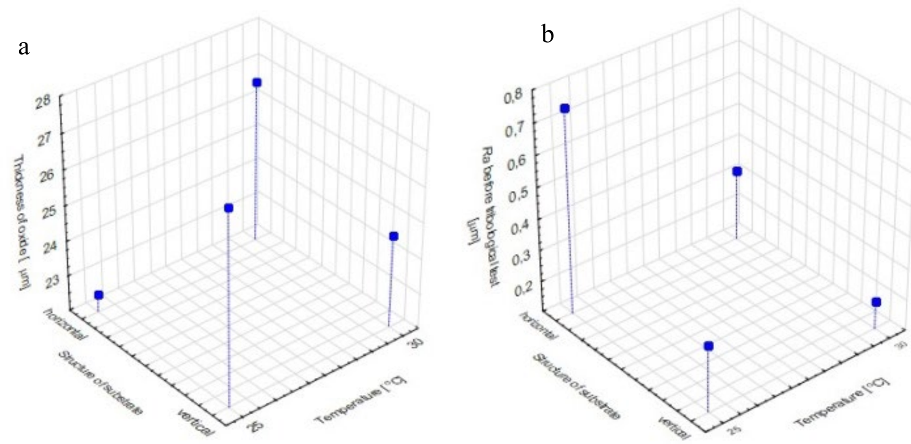


Figure 1. *The scatter chart of **(a)** oxide thickness and **(b)** Ra parameter before the tribological test over the structure of substrate and temperature of anodizing process.*

Table 2. A rule of thumb scale for evaluating the correlation coefficient

Strength of Correlation	
Size of r	**Interpretation**
0.90 to 1.00 (-0.90 to -1.00)	Very high positive (negative) correlation
0.70 to 0.89 (-0.70 to -0.89)	High positive (negative) correlation
0.50 to 0.69 (-0.50 to -0.69)	Moderate positive (negative) correlation
0.30 to 0.49 (-0.30 to -0.49)	Low positive (negative) correlation
0.00 to 0.29 (0.00 to - 0.29)	Little if any positive (negative) correlation

Table 3 The values of correlation coefficient between the analyzed variables.

Variables \ Variables	**Strength of Pearson's correlation coefficient *r***			
	Friction coefficient	**Wear intensity of PEEK/BG**	**Thickness of oxide**	***Ra* parameter**
Thickness of oxide	-0.15	0.33	-	-
Temperature	-0.99	-0.87	0.17	-0.64
Structure of substrate	-0.003	-0.24	-0.38	0.68
***Ra* parameter**	0.64	0.25	-	-
Wear intensity of PEEK/BG	0.88	-	-	-

Pearson's correlation coefficient (Table 3) between the wear intensity of PEEK/BG plastic and temperature was r = - 0.87, which means a high negative correlation. Such variables as thickness of oxide, structure of substrate and Ra roughness parameter of oxide layers show low or no correlation.

Table 4 shows the pictures of surface coatings and isometric projection of surface before and after tribological tests. The picture of surface coating before and after tribological test depicted the type of a surface substrate which was called in this article "horizontal" for samples whose surface addicted by the production of aluminum alloy was parallel to the direction of motion in tribological tests and called "vertical" for perpendicular direction of motion. The isometric projections clearly show how the roughness of surface is reduced after tribological tests in comparison with the value before tribological test.

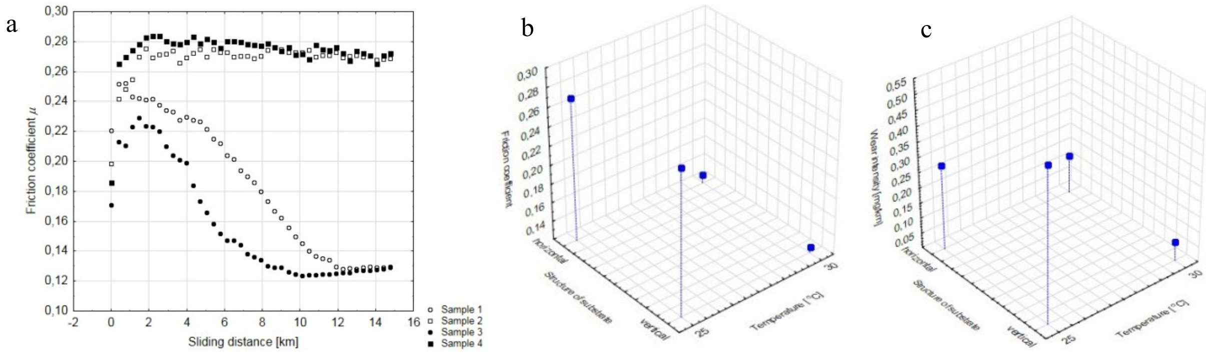
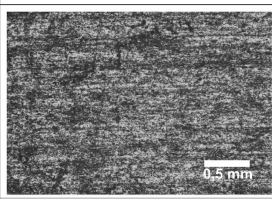
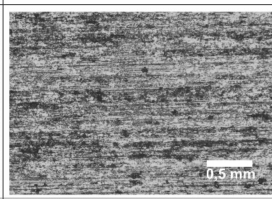
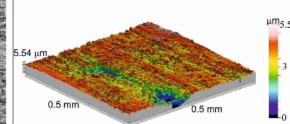
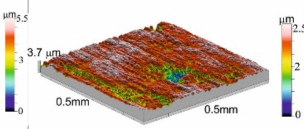
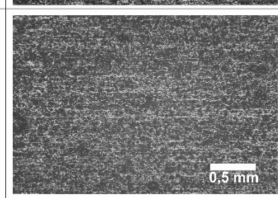
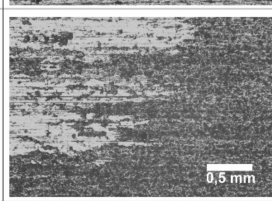
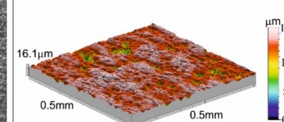
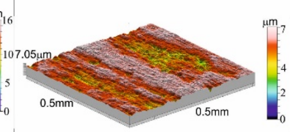
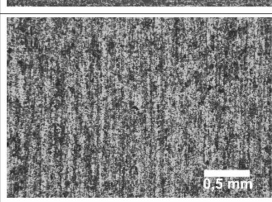
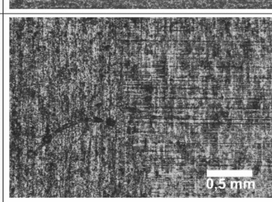
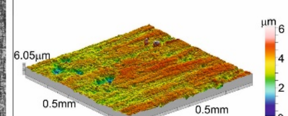
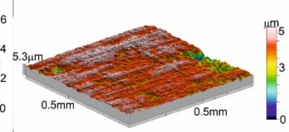
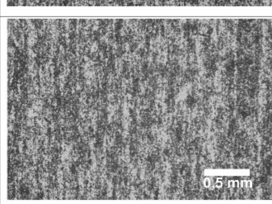
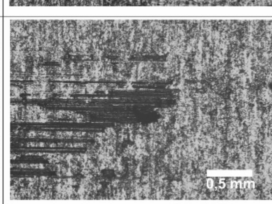
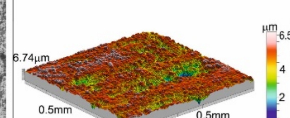
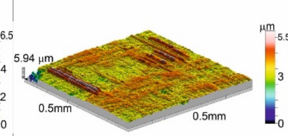


***Figure 2.** The diagram of friction coefficient versus sliding distance **(a)**, the scatter chart of friction coefficient μ **(b)** and wear intensity of PEEK/BG plastic **(c)** over temperature of anodizing process and structure of substrate.*

Table. 4. Pictures of surface coatings and isometric projection of surface before and after tribological tests.

Sample name	Before tribological test	After tribological test	Before tribological test	After tribological test
1				
2				
3				
4				

Conclussion

This paper gives an account of the 2k factorial design which was used to determine the influence of electrolysis temperature and the structure of a substrate on the tribological properties of Al_2O_3/IF-WS_2 coatings. Taken together, these studies indicate that the structure of an aluminium substrate has no influence on the friction coefficient from the point of view of the orientation of samples during a tribological test. The connection of design of experiment and tribological experiment emphasizes the validity of obtained results. The lower values of friction coefficient μ and wear intensity of PEEK/BG plastic during abrasive wear were obtained for samples obtained in the higher temperature of 30°C. The analysis of Pearson's coefficient confirms that the friction coefficient μ and wear intensity of PEEK/BG plastic depend on the temperature of electrolyte during electrolysis and surface roughness parameter *Ra*.

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