EFFECT OF MICROSTRUCTURE AND MECHANICAL PROPERTIES ON WEAR RESISTANCE OF SILICON NITRIDE CERAMICS

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Abstract

The wear of Si_3N_4 ceramics with high-resistant abrasive material during friction applications was analyzed. Effects of the hot pressing parameters on the microstructures and properties of ceramic samples were evaluated. Negative influence of grain growth with increasing of the β-Si_3N_4 phase ratio on wear resistance of tested materials was proved. The volume loss of separate ceramic materials during the wear tests depended mainly inversely proportional on the hardness of ceramics. Both microcutting and microcracking mechanisms took part during the wear of Si_3N_4 ceramics.

Keywords: *ceramics, silicon nitride, microstructure, wear resistance*

1. Introduction

In the past few years, silicon nitride ceramics has been increasingly used as wear resistant material. High-temperature strength and hardness at acceptable toughness, good chemical stability and excellent thermal shock resistance are considered as the most important properties of those ceramic materials [1,2]. Silicon nitride has been successfully used in a wide variety of engineering applications involving contacts with metallic surfaces such as drawing dies, roller bearings, cutting tools, automotive or aerospace engine parts [3-5].

Many studies, which are devoted to problems of ceramic wear resistance, confirmed the relationship between wear and some working parameters such as load, sliding and speed or kind of wear [2-5]. Abrasive wear is the most common mechanism of ceramic material removal. Two basic mechanisms of surface damage are applied during the abrasive wear of ceramics: microcutting and microcracking [6].

The mechanisms of microcutting, is simplified as illustrated in Fig.1, where the abrasive particle in the cone shape grooves the solid body surface. This model comes from Rabinowitz´s conception of abrasive wear mechanism description [7]. According to this assumption, the force F is necessary to get the cone into a depth h. The value of this force can be calculated according to equation (1).

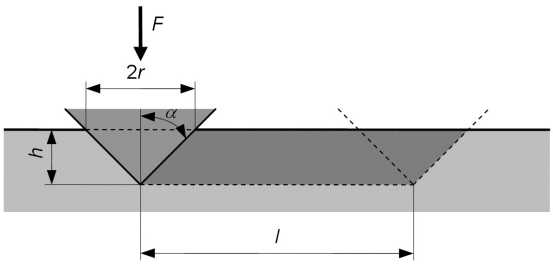

Fig. 1. Model of microcutting material surface by an abrasive grain

$$F = H_V.\pi.r^2 \tag{1}$$

In this equation, H_V is the hardness of worn material and r is radius of impression. If the cone moves during relative motion in parallel with the worn material surface, it creates a groove on the surface with length l and it removes volume V, which is described using equation (2).

$$V = h^2.tg\alpha.l \tag{2}$$

In equation (2) α is the angle of cone which participates in the grooving of the surface. By substituting r with $h.tg\alpha$ in equation (1) and replacing h in equation (2) the removed material volume V can be described by equation (3).

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$$V = \frac{F.l}{\pi.H_V.tg\alpha} \tag{3}$$

It follows from equation (3) that the removed material volume is dependent only on one material property, hardness H_V.

The mechanism of microcracking is possibly explained by means of Fig. 2. Normal stress, which effects the abrasive particle, creates the area of plastic deformation under a solid figure surface. This effect creates a crack in the plane of the load axis after overrun of the specific limit value of the load. This crack is called median and stably grows by increasing the load. During the unloading phase, the median crack is closed and lateral cracks are created in the area of plastic deformation in consequence of residual strains (Fig. 2). After full removal of external normal stress (represented by force F) the lateral cracks propagate further to the sample surface, where it can come to fracture.

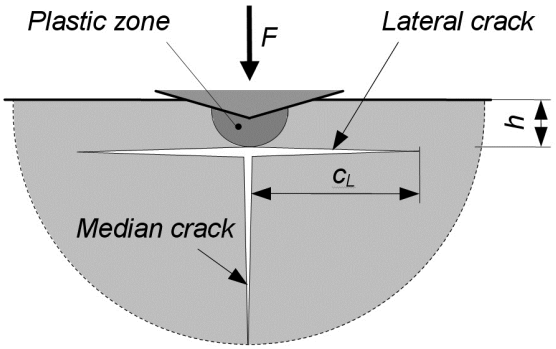

Fig. 2. Model of microcracking material surface by an abrasive grain

Volume of wear V is possible to express by means of equation (4).

$$V = h.c_L.l \tag{4}$$

In this equation (4) h is depth in which the crack occurs, c_L is length of a lateral crack and l is length of path of abrasive particle. The length of the lateral crack can be expressed according the equation (5). The depth of the crack can be calculeted by equation (6).

$$c_L = \frac{F^{5/8}}{K_{IC}^{1/2}.H_V^{1/8}}.\left(\frac{E}{H_V}\right)^{2/5} \tag{5}$$

$$h = \left(\frac{F}{H_V}\right)^{1/2}.\left(\frac{E}{H_V}\right)^{2/5} \tag{6}$$

where F is load on abrasive particle, K_{IC} is fracture toughness of ceramics, H_V is hardness of ceramics and E is modulus of elasticity of worn ceramic material.

After substitution of equations (5) and (6) into equation (4) the final equation (7) is valid for the calculation of removed material volume [2, 8].

$$V = \frac{F^{9/8}}{K_{IC}^{1/2}.H_V^{5/8}}.\left(\frac{E}{H_V}\right)^{4/5}.l \tag{7}$$

According to equation (7) the removed material volume V is dependent on three materials properties: modulus of elasticity E, fracture toughness K_{IC} and hardness H_V.

According to some works [7, 9] the ratio K_{IC}/H_V values appears as the main characteristic determining dominant wear mechanisms of brittle materials during abrasive wear. This parameter defines dominant wear mechanism in contact. The mechanism of plastic microcutting dominants at a high value of this rate, i.e. wear volume will be depended on hardness ($V \sim 1/H_V$). Brittle fractures dominant at a low value of the rate K_{IC}/H_V, i.e. wear will increase with decreasing fracture toughness ($V \sim K_{IC}/H_V$). This influences growth of wear intensity. Acoording to equations (3) and (7) the intensities of both microcutting and microcracking increase, when we increase the load on abrasive particle during the wear. To the contrary the intensity of microcutting decreases with the hardness increasing, and the intensity of microcracking decreases with fracture toughness of the worn surface. This can lead to a transition from plastic microcutting to brittle microcracking during abrasive wear [10].

One possibility how to achieve high wear resistance is to obtain a homogenous fine grained structure. For this reason, the character of structure plays an important task at determinating how ceramics will react to specific states of stress which arises in specific conditions of wear. Grain size is an important factor of tribological properties of ceramic material determination [3, 4, 11].

2. Description of materials and experiments

The ceramic material chosen in this study was silicon nitride hot-pressed with sintering additives of yttrium oxide and aluminum oxide. The amounts of Y_2O_3 and Al_2O_3 sintering additives in separate specimens were different but the proportion of Y_2O_3 and Al_2O_3 was the same for all prepared specimens. The Y_2O_3 and Al_2O_3 powders were added in concentrations that can set creating of $Y_3Al_5O_{12}$ phase in ceramic materials. This phase contributed to the sintering ability of ceramics [12]. The designations and compositions of studied materials are given in table 1. The total amount of sintering additives varied from 10 to 15 wt. %.

Tab.1

Chemical composition (in wt. %) of prepared ceramic materials

Ceramics	Si_3N_4	Y_2O_3	Al_2O_3
SN10	bulk	7.75	2.14
SN12	bulk	8.78	3.00
SN15	bulk	10.34	4.30

Initial powder compositions were wet mixed in alcohol. After drying and sieving, the powder was compacted in steel dies. The final densification was accomplished using hot pressing techniques in a nitrogen atmosphere with the purity of 99.99 % and an overpressure of 75 kPa. All samples were hot pressed at a temperature of 1680 °C and a pressure of 34 MPa. Three various sintering times 5, 15 and 30 min were applied. Ten samples were pressed at every composition and sintering times, thus 90 samples were prepared. All samples had diameters of 8.4 mm and a height of 10 mm.

Densities of hot pressed ceramics were measured by the Archimedes's method. Hardness was determined by means of Vickers indentation method with a testing load of 98 N. The fracture toughness was also determined by means of Vickers indentation. This method is based on the measurement of crack lengths that rise from the indentation corners. The more brittle the tested material is, the longer are initiated cracks.

The wear resistance was evaluated by means of grinding the sample using pin on disk method. Test samples were placed in contact with corundum grinding paper with a graininess of 120 μm. The grinding trajectory was 125 m and pressure 1.5 MPa. The wear resistance was determined based on the volume loss of the samples related to the grinding trajectory according to equation (8)

$$V = \frac{\Delta m}{\rho . l} \tag{8}$$

where V is volume loss of the samples, Δm is weight loss of the samples, ρ is density of the sample and l is grinding path of the sample.

Microstructures of hot pressed ceramics with different contents of sintering additives and different pressing time were observed using a scanning electron microscopy. In order to identify the created microstructure, the hot pressed specimens were subjected to XRD analysis.

3. Description of achieved results

The effects of physical and mechanical properties on the microstructure and wear resistance of the prepared ceramics samples were evaluated.

3.1. Physical properties of ceramics

Average values from ten measured densities are included in table 2. The samples with every different compositions and pressing time were analyzed on the phase composition by XRD method (tab. 3). Only two phases were found in all specimens α-Si_3N_4 and β-Si_3N_4 phases. Phases with a con-centration below 5 % couldn't be identified by used the XRD method.

Tab.2

Density of ceramics sintered for different times

Ceramics	Density ρ (g.cm^{-3})		
	5 min	15 min	30 min
SN10	3.21	3.24	3.25
SN12	3.23	3.27	3.26
SN15	3.27	3.27	3.26

Tab.3

β-Si_3N_4 phase ratio of ceramics sintered for different times

Ceramics	β-Si_3N_4 phase ratio (wt.%)		
	5 min	15 min	30 min
SN10	45	58	75
SN12	42	69	81
SN15	43	62	100

The densities of ceramic samples were influenced by the amount of additives (Y_2O_3 and Al_2O_3) and by the sintering time. They increased with increasing the concentration of additives and sintering time from 3.21 to 3.27 g.cm^{-3}. These values corresponded to the relative density from 97.2 to 98.5 %, which indicate good densification of samples. The smallest density 3.21 g.cm^{-3} was measured at the sample SN10 with the smallest concentration of the additives sintered for 5 min. The highest value of 3.27 g.cm^{-3} was achieved at sample SN12 sintered for 15 min and at sample SN15 sintered for 5 and 15 min.

During hot-pressing, the initial α-Si_3N_4 powder was transformed to β-Si_3N_4 phase, which can be seen from table 3. The transformation stage also increases with both the additive concentrations and the sintering time. The portion of β-Si_3N_4 phase from 42 to 45 % was measured by XRD at a shorter sintering time of 5 min of all prepared compositions. There were small differences between separate compositions, but these were within the measurement error, which was 5 %. Whole 100 % transformation of α-Si_3N_4 powder to β-Si_3N_4 phase was achieved only at samples SN15 with the highest concentration of additives and sintered for 30 min.

3.2. Microstructure of ceramics

The study of microstructure confirmed the effect of the composition on the formed phases, which can be seen in Fig. 3, 4 and 6, where we can see the ceramics with various contents of additives but the same sintering time of 30 min. The amount of overextended β-Si_3N_4 grain increased with the additive content. That can be explained by the increasing of transformation velocity at higher additive contents. With the increasing of the additive amount the ratio of a binding phase at the grain boundary increased. This crystal boundary phase is relatively brittle and can by indicated as the weakest component of the microstructure [12-14].

The grain size comparison for samples SN12 with the middle concentration of additives shows that the grains grew with the sintering time excessively (Fig. 5 and 6). Similar effect was also observed at other compositions.

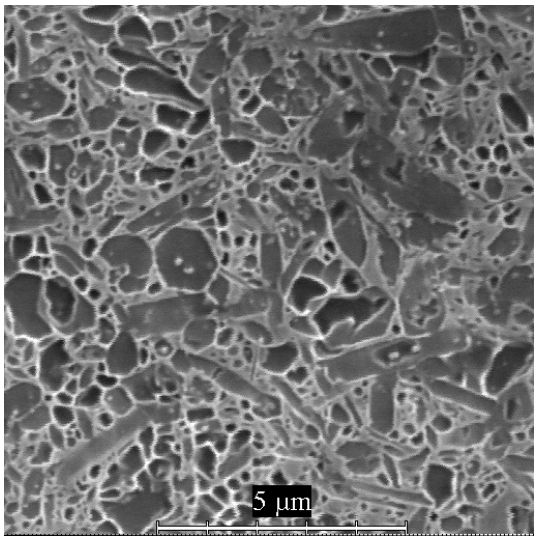

Fig. 4. Microstructure of ceramics SN15 sintered for 30 min

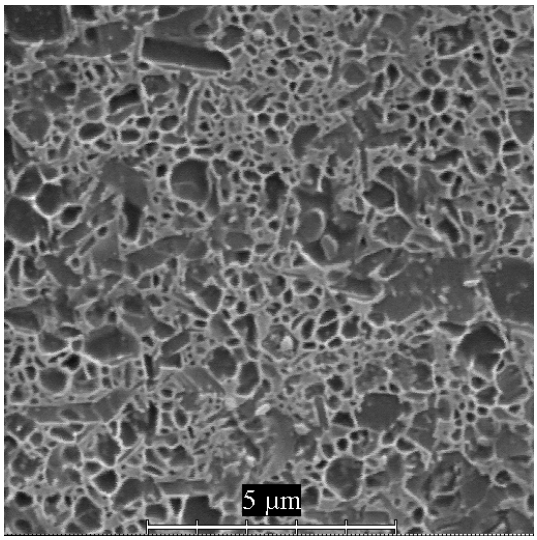

Fig. 5. Microstructure of ceramics SN12 sintered for 5 min

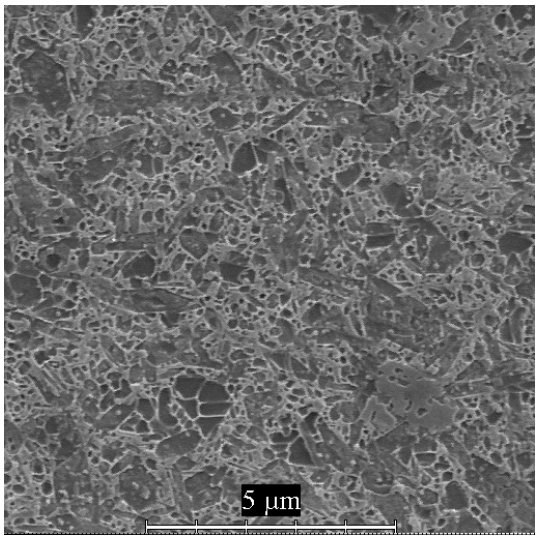

Fig. 3. Microstructure of ceramics SN10 sintered for 30 min

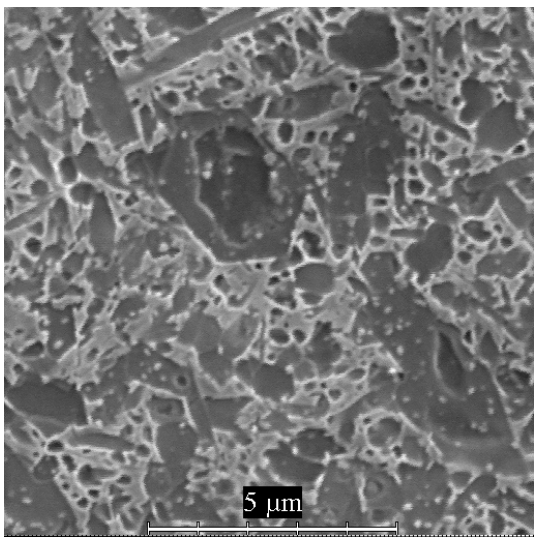

Fig. 6. Microstructure of ceramics SN12 sintered for 30 min

3.3. Effects of sintering parameters and mechanical properties on wear

The effect of addition concentration and sintering time on the wear of ceramic samples is in Fig. 7.

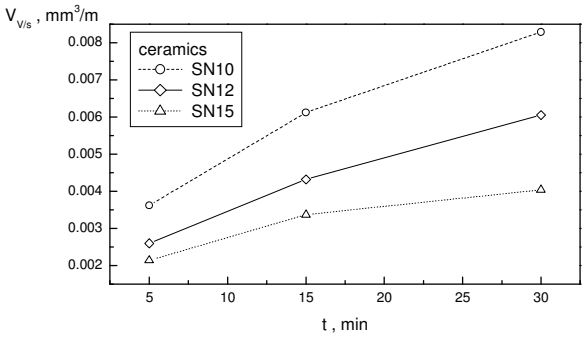

Fig. 7. Effect of pressing time and additive concentration on wear of ceramics

From this figure it can be seen that the volume change during the wear test decreased with increasing of additions content and increased with sintering time. The highest wear resistance was achieved at the sample SN15, which was pressed at only 5 min, and the least wear resistance was with the specimen SN10 with the smallest addition content pressed for the longest time of 30 mins.

The effects of measured mechanical properties on the wear properties of ceramic samples were studied in detail and are presented in the Fig. 8, 9 and 10.

Hardness had the positive effect on the wear resistance (Fig. 8). Higher hardness resulted in less wear. As the highest hardness was measured at the higher concentration of additives (ceramics SN15), these samples had the smallest volume wear changes. The highest volume wear changes were measured at samples SN10 with only 10 wt.% of additives that was pressed for 5 min. The results in Fig. 8 are in good correspondence with model ($V \sim H_V^{-1}$), when the volume losses V during the wear tests depend inversely proportional on hardness H_V of ceramics.

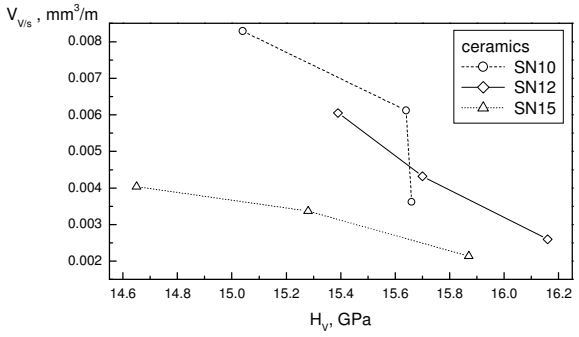

Fig. 8. Effect of hardness on wear of ceramics

A very interesting development was measured with the effect of fracture toughness on wear in Fig. 9. All compositions had the same progress. At first the wear increased to the maximum value. After reaching the maximum value the wear slightly decreased. This can be explained by the relationship between the β-Si_3N_4 phase and grain size, but the differences between the separate samples within each compositions were relatively small. The effect of grain size was dominant. The highest grain size was always at the sample with a middle value of fracture toughness. That means the wear behavior can't be described only by the effect of fracture toughness on wear.

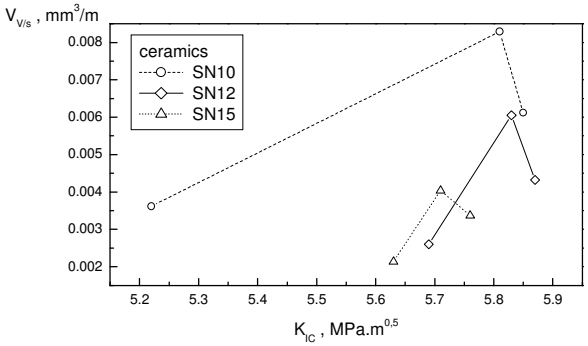

Fig. 9. Effect of fracture toughness on wear of ceramics

Wear behavior can be sometimes better described by the model which reflects the effect of ratio fracture toughness / hardness on wear rate. This can be seen in Fig. 10.

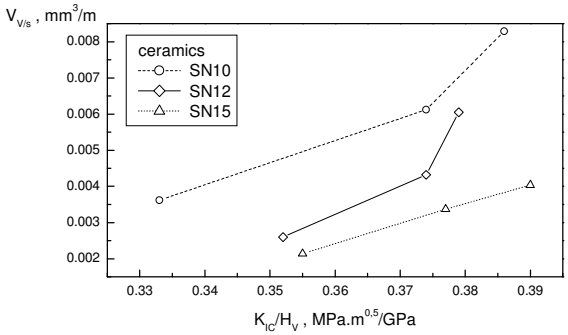

Fig. 10. Effect of ratio K_{IC}/H_V on wear of ceramics

These relations fit very well for every separate composition. The higher the value of calculated ratio of fracture toughness to hardness, the higher is the wear rate. The highest wear rate was noticed with the specimen SN10 that contained the smallest concentration of additives that was pressed for 30 min. The ratio of fracture toughness to hardness very well describes the relationship between the decreasing of wear resistance in spite of the transformation progress α-Si_3N_4 phase to the β-Si_3N_4 phase and increasing of wear rate in spite of grain growth.

3.4. Ceramics surface after the wear test

Specimen surfaces with different content of additives sintered for 5 min after the wear tests are documented in the next figures 11 and 12. Fig 13 and 14 show ceramic surface with the middle contents of additives sintered for different times 5 and 30 min. These figures confirmed the effect of additive concentration and sintering time on the wear resistance of specimens that were described above. We can see that the specimens which were pressed for a longer time (30 min) showed larger damage than the specimens pressed for a shorter time (5 min) (figure 13 and 14). The specimens with higher contents of additives (in Fig. 12) showed smaller damage than the specimen with smaller additive concentrations in Fig. 11. These specimens have higher portions of binding phases, which is present on the grain boundary. The binding phase has a positive effect on the densification, which generally improves the mechanical properties of ceramic materials. On the other hand the binding phase is the weakest component of the microstructure. This brittle crystal boundary phase broke and chipped off during the wear tests, which can be seen in Fig 14. Prolongation of pressing time is joined with grain growth, which caused less wear resistants, thus the larger volume of the surface were enucleated.

Consistent with theoretical background, the volume loss depends on hardness ($V \sim H_V^{-1}$) when the dominant mechanism of wear is microcutting. If the dominate mechanism is microcracking, the volume loss depends on the ratio hardness / fracture toughness ($V \sim K_{IC}/H_V$). All ceramic surfaces were damaged by microcutting and also by microcracking. This can be clearly seen from the observed surface after the wear tests. For example in Fig. 13 and 14, which revealed the specimens with middle concentration of additives and pressed for different times, several scratches and also damaged areas are visible. These areas are characteristic with dropped out material and many microcracks. These characteristic scratches and pits with microcracks are presented on all specimens. Thus both mechanisms took part of wear of all specimens. However the extent of ceramic surface damage increased with pressing time.

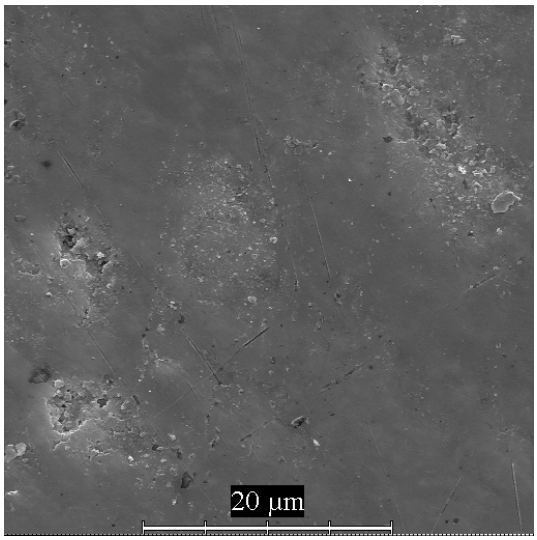

Fig. 12. Surface of ceramics SN15 sintered for 5 min after wear test

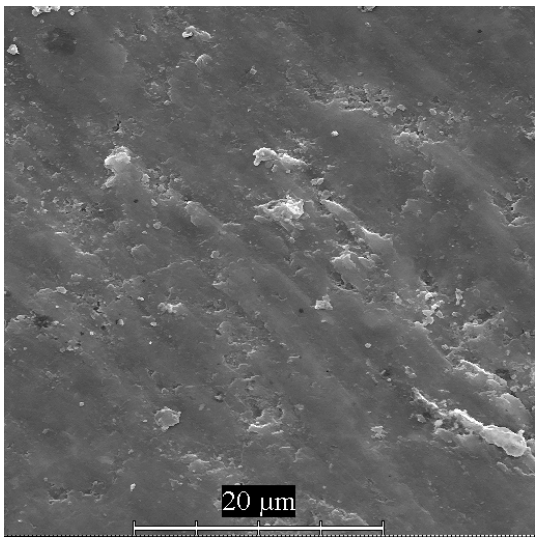

Fig. 11. Surface of ceramics SN10 sintered for 5 min after wear test

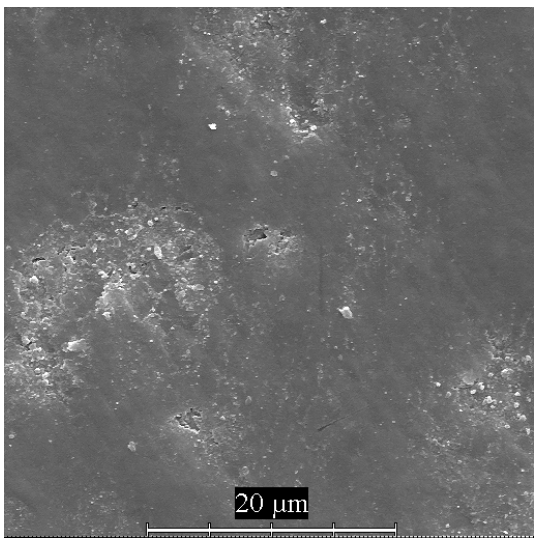

Fig. 13. Surface of ceramics SN12 sintered for 5min after wear test

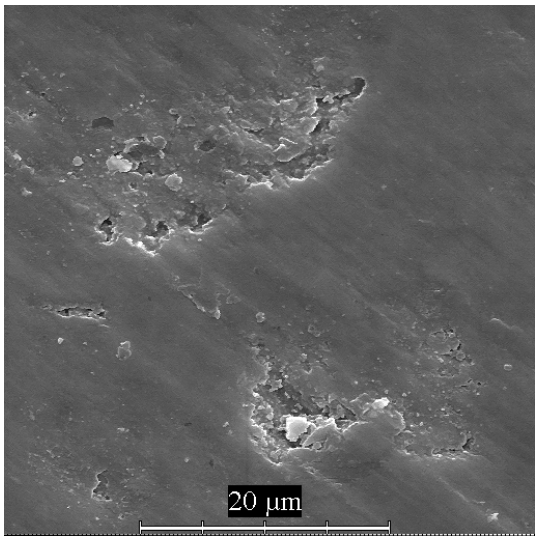

Fig. 14. Surface of ceramics SN12 sintered for 30 min after wear test

In the case when both mechanisms took place during the wear of Si_3N_4 ceramic with alumina abrasive, both models could be used to interpret experimental dates. In our case, the experimental data were in better correspondence with the model, which assumes that microcutting as the dominant mechanism. Since there were only very small changes in the fracture toughness of separate specimens, the effect of ceramic hardness can be consider as dominant. Thus the volume loss during wear can be described by the first model ($V \sim H_V^{-1}$).

4. Conclusions

This paper concentrated on the analysis of the wear mechanism of silicon nitride based ceramics, which was prepared by hot pressing techniques. The effect of chosen preparation parameters like chemical composition, sintering conditions on microstructure, mechanical properties and wear resistance was evaluated.

Wear was mostly influenced by the hardness of ceramic materials. The specimen with the highest hardness achieved the highest wear resistance. Wear resistance of ceramics decreased with the grain growth and with the transformation progress of narrow α-Si_3N_4 phase to prismatic β-Si_3N_4 phase. The weakest microstructure constituent was the brittle crystal boundary phase that broke and chipped off during the wear tests.

Two damage mechanisms were taken into consideration on ceramic surfaces during the wear of Si_3N_4 ceramic pin samples with alumina grinding disks. Specimen surfaces were damaged by microcutting and also by microcracking mechanism. The volume loss V of separate ceramic materials during the wear tests depended mainly inversely proportional on the hardness H_V of ceramics. Based on these experimental results the volume loss and also the wear resistance of studied ceramics could be described by model $V \sim H_V^{-1}$.

Acknowledgements

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