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Effect of hybrid SiC/TiO_2 nanoparticles on tribological and mechanical performance of polymethylmethacrylate dental base material

ABSTRACT

Many researches dealt with PMMA dental base material and the development of its properties to reach a longer life span for implants and fillings. The current work pay attention to examine the loading amount of hybrid nanoparticles, SiC and TiO_2, and find out how it affects the mechanical and tribological properties. Hybrid NPs were dispersed on PMMA resin with filler amount of 0.4%, 0.8%, 1.2%, 1.6%, and 2.0 wt.%, 50/50 between SiC and TiO_2 NPs. The mechanical properties were evaluated by determining the hardness, Shore D, compressive strength, and modulus of elasticity. While the tribological performance was assessed via examining the COF, wear rate and scanning the worn surfaces using optical and SEM images. The results can be indicated that the PMMA resin exhibits a good reaction bonding with low loading amount of the hybrid NPs. Moreover, the high loading content had a negative effect on the mechanical and tribological properties. Subsequently, the loading content of 0.8 wt.% of SiC/TiO_2 NPs indicates that it has the best performance comparing with the pure PMMA.

Keywords: *PMMA, Friction, Wear rate, Mechanical properties, SiC Nano Particles, TiO_2 Nano Particles.*

1. INTRODUCTION

Polymers are unique in that they are huge numbers of smaller molecules that bond chemically among themselves to form macromolecules, which facilitates the control of the physical, chemical, and mechanical properties of the bulk polymer [1]. These distinctive features made polymers one of the most widespread and developed materials in various fields. Several studies dealt with and sought to develop and improve the properties of polymers to suit the specific application [2–11]. Polymers are characterized by being a biomaterial, which imposed them as an Restorative material for hip joints, bones and dentistry [12–18]. Amalgam is labeled as one of the first restorative material has been used in dentistry, as it began to be adopted over a period of 150 years [19,20]. Composite resins were approved as a dental restorative material for more than 100 years. Since then, much research has begun to study the properties of the resin and work to improve its properties [21-23].

Polymethylmethacrylate (PMMA) is a durable, transparent, available, biocompatible, and safe polymeric material [24]. Due to its excellent physical, mechanical, tribological and esthetic properties, PMMA has been widely used in dentistry, as fillers, implants, and prosthesis. PMMA spherical particles can be produced via alginate or gelatin stabilizer techniques [25,26]. Recently, several studies have demonstrated the development and improvement of mechanical and physical properties when incorporating different nanoparticles (NPs) into a polymer matrix. Many natural materials were incorporated to PMMA to improve the mechanical and tribological performance of the resin. Miswak and corn cobs particles were inhabited a good wear resistance and antimicrobial effect [27–29]. Multi-walled (MWCNTs) and single-walled (SWCNTs) carbon nanotubes were utilized as filler to PMMA dental base material. Flexural strength of PMMA resin reinforced with 0.5, 1.0 and 2.0 wt. % content of MWCNTs, were analyzed and the PMMA nanocomposites display a good improve on the mechanical properties [30]. The tribological and mechanical performance of samples of PMMA with low loading amount, 0.1, 0.2, 0.3, 0.4 and 0.5 wt. %, were examined. PMMA/0.3% MWCNTs exhibits a good attitude on friction and wear rate comparing

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with the pure PMMA [16,17]. Also, the hardness of PMMA reinforced with CNTs was investigated [31]. Moreover, PMMA hot acrylic reinforced with SWCNTs was evaluated against various counter faces, stainless steel, porcelain, amalgam, and buffalo teeth, under dry and wet conditions [32]. Carbon and graphite fiber was used to develop the mechanical characteristics of PMMA resin. The PMMA composites showed an amelioration of Fracture stress and flexural modulus [33]. Also, the PMMA dispersed with hydroxyapatite with small filler amount, has a direct impact on distinguishing mechanical and tribological performance [34].

Several studies were evaluated to explore the effectiveness of dispersion of various metal oxide NPs through PMMA acrylic on the mechanical, physical properties and wear resistance. Alumina with various forms as spherical NPs, and an electrospun fibers were used as fillers to PMMA matrix. Sample PMMA which filled with 3 wt. % of electrospun Alumina, exhibits an increase in hardness up to 157.8% compared to the pure PMMA [35]. PMMA/Al_2O_3 high loading filler amount of 5, 10 and 15 wt.% were evaluated using compression molding technique. It was noticed that the modulus of elasticity increases with 25% for PMMA matrix loading with 15 wt. % of Al_2O_3 [36]. Effect of a low content of Al_2O_3 NPs dispersion on PMMA based resin was performed. For the results, it can be observed that the best tribological performance were obtained at a loading amount of 0.6 wt. % [37]. TiO_2 NPs is distinguished by some attributes that qualify it as an optimal filler. Where it is a biocompatible, antibacterial, good corrosion resistant, high durability, bonding stable and inexpensive [38]. The mechanical performance and homogeneous of PMMA/TiO_2 nanocomposites proved to be an outstanding restoration material [39]. Methacrylic acid may be used as a coupling agent to produce PMMA/TiO_2 nanocomposites. Elasticity, shear modulus and thermal analysis of the matrix were evaluated. It was found that, transition and degradation temperatures increase due to increase of TiO_2 content [40]. Graphene oxide (GO) was added to PMMA/TiO_2 to analysis the mechanical, thermal and microstructure demeanor. It can reveal from the results that GO play a prominent role to improve the properties of PMMA dental resin [41]. Hybrid SiO2/TiO_2 was dispersed via PMMA resin to examine its properties. Nano-graphene was added to the matrix to benefit from self-lubrication to reduce friction [42].Silicon carbide NPs was evaluated to coat the titanium implants using Plasma-enhanced chemical vapor deposition (PECVD). It can be concluded that titanium implants coated with SiC exhibit a harmonious distribution and durability under torque upon implantation [43]. Thermal conductivity and flexural strength of PMMA matrix were evaluated with loading content of 10 wt. %. From this study, it was observed that Al_2O_3 and SiC filler powders increase the thermal conductivity of PMMA resin with no negative effect on its strength [44].

The aim of this work is focused on investigate the role of adding SiC NPs to TiO_2 NPs on the mechanical and tribological performance of PMMA base dental material. The friction coefficient, wear rate and worn surface were performed to evaluate the tribological characteristics, while hardness, XRD, and modulus of elasticity were analyzed to assess the mechanical properties.

2. MATERIALS AND EXPERIMENT SETUP

2.1. Materials and Samples Preparation's

PMMA acrylic resin is the base dental material that was adopted in this work. PMMA acrylic, purchased from Acrostone Co. - Egypt, is a cold cure resin. Commercial acrylic is a two-component phrase, one being solid, PMMA powder, and the other being liquid, called monomer MMA. The filler materials used in this research are TiO_2 NPs and SiC NPs, were supplied by US Research Nanoparticles Inc. Table 1 illustrates the attributes of TiO_2 NPs and SiC NPs.

Table 1. Technical Properties of TiO_2 NPs and SiC NPs

Tabela 1. Tehnička svojstva TiO_2 NPs i SiC NPs

Description	TiO_2	SiC
Purity %	99.5	99
Color	white	black
Average Particle Size [nm]	40-50	18
Specific Surface Area [m^2/g]	480-650	80-135
Bulk Density [gm/cm^3]	0.12	0.03
True Density [gm/cm^3]	3.9	3.216

PMMA matrix produces by blending both PMMA powder and MMA liquid in a 2:1 ratio. Polymerization process between the monomers and the powder acts through a reaction between the matrix molecules, which produces a solid mass. Samples were prepared in a container by dispersing the nanoparticles in the PMMA matrix using a rotary stir with 300 rpm of 15 min, at 30° C and humidity 40%. As the stirring continues, the mixture gradually begins to thicken due to the partial dissolution of the resin particles and turns into a dough. The mixture was poured into a cylindrical mold, with dimensions of 20 mm x 10 mm, and pressurized at 5 bars for 1 hour, then left for a week in the open air to complete hardening. Sample set is prepared with filler loading amount of 0, 0.4, 0.8, 1.2, 1.6 and 2.0 % of weight of resin. Both of TiO_2 and SiC NPs are calculated in equal proportions. Figure 1 displays steps of the sample preparation.

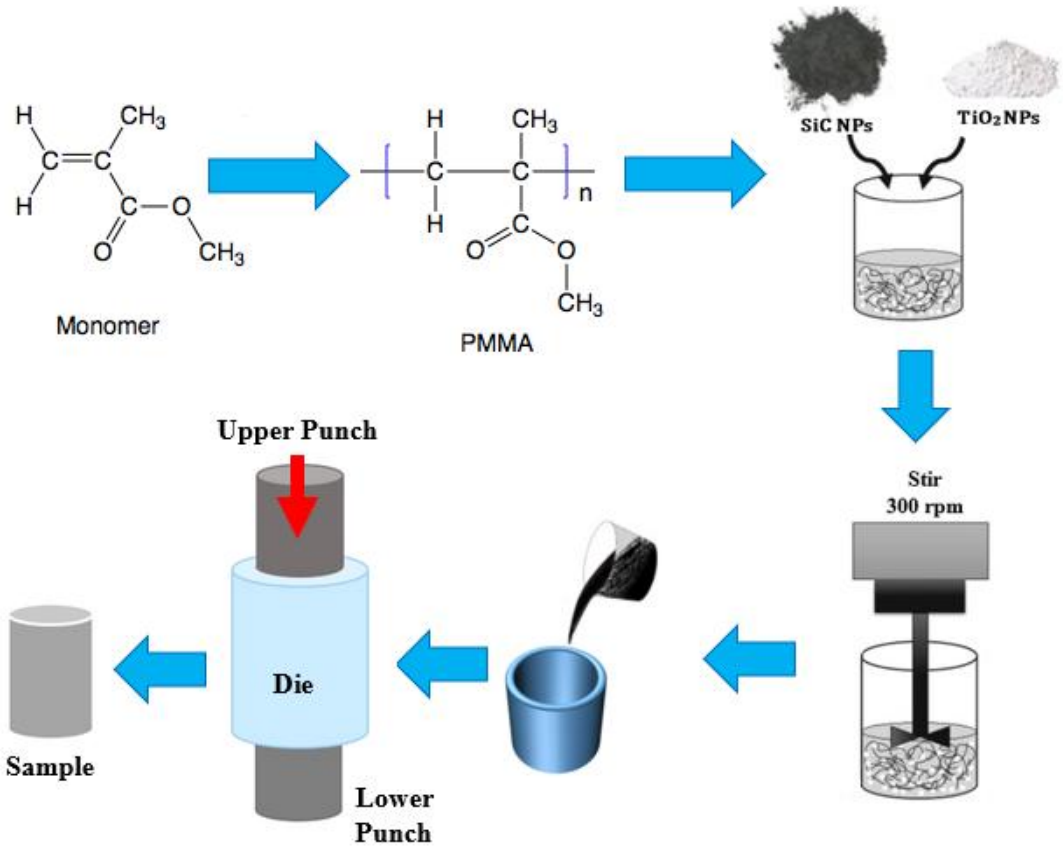

Figure 1. Schematic steps of the PMMA samples' preparation

Slika 1. Šematski prikaz pripreme PMMA uzoraka

2.2. Experimental Setup

a. XRD Pattern

X-Ray diffraction analysis (XRD) was performed by using X-Ray Diffraction (Shimadzu 6000). Microstructural analysis confirmed to identify of crystallinity with K_α ray at high-speed rate of 1000 /min and high-precision angle reproducibility of 0.001°. The data were collected at independent dual axis θ.

b. Mechanical Properties

The mechanical properties were evaluated via durometer Shore D equipment and United Universal Hydraulic (DFM-300KN). According to ASTM standard D2240 [45], the hardness value was confirmed and calculated the average of the values for five points along the sample surface. The compressive strength was measured via confirming the compression load carrying capacity, based on ASTM standard D1621 [46].

c. Tribological Properties

The tribological performance was assessed via a reciprocating tribometer, pin-on-disk, based on ASTM G99-95a [47]. The test rig illustrates in Figure 2. The friction coefficient and wear rate were evaluated under operating conditions of 30°C, a humidity of 40% and dry contact. The experiments were conducted under various applied loads of 4, 6, 8, 10 and 12 N at a linear velocity of 0.4 m/s. The samples were examined as pin slides against a rectangular disk made of stainless steel. Disk surface is carefully cleaned before each experiment with acetone to remove any contaminants and then surface dried with a heat gun. The sample is weighed before and after the experiment to determine the weight loss and to evaluate the wear rate. To avoid an error, the average values of the wear rate and the friction coefficient are calculated by performing the test for each sample five times under the same conditions.

d. SEM Analysis

The worn surfaces of samples were evaluated using SEM microscope (JCM-6000Plus; JEOL, Tokyo, Japan). The detailed description of samples surfaces was achieved via preparing the surfaces. The surfaces are cleaned and air dried, then covered with a thin film of platinum after that the images are taken.

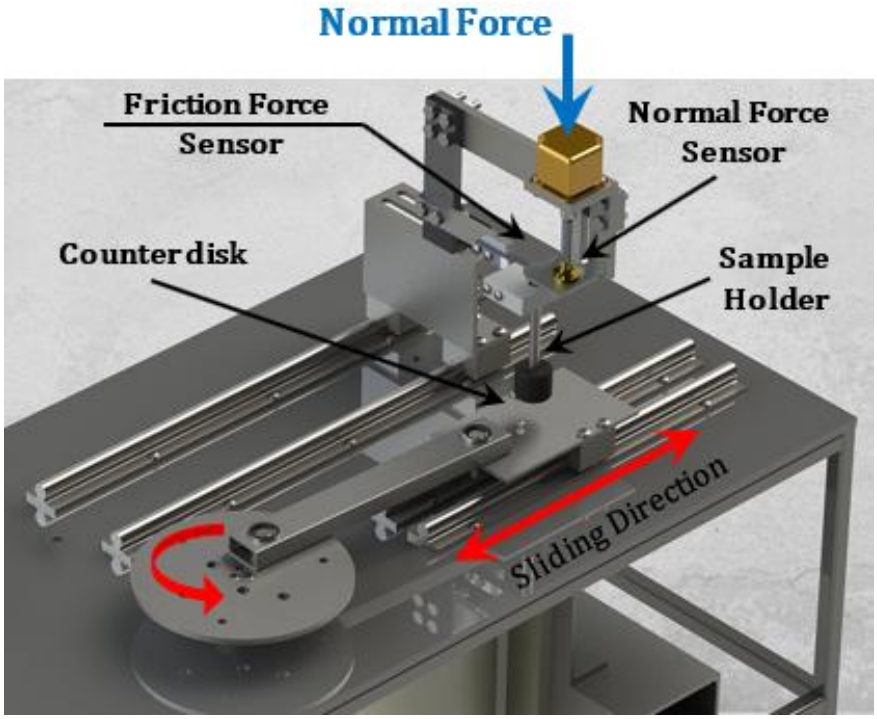

Figure 2. A reciprocating pin-on-disk tribometer

Slika 2. Klipni tribometar sa pin-on-disk

3. RESULTS AND DISCUSSION

Typical X-ray diffraction patterns were performed to assess crystallinity and phases of the products. Figure 3 illustrates the XRD pattern of SiC and TiO_2 NPs. It can be observed that SiC spectrum appears four diffraction peak points existed at 2θ = 35°, 41°, 60°, and 72° [48]. While it can be noticed that several diffractions centered at 2θ = 25°, 39°, 48°, 55°, and 63° on TiO_2 NPs spectrum XRD [49]. It can be deduced that the SiC and TiO_2 NPs have a good crystalline and distinctive purity due to the absence of peaks indicating that it contains impurities [50,51]. Figure 4 illustrates XRD spectrum for PMMA samples with different loading content. For pure sample spectrum, it can be found that strong intensity peak centered at 2θ = 14.5˚. Moreover, the 2θ peaks at 28.5˚ and 41.5˚ matches with previous research [52]. As nanocomposite samples show a positive pattern, which gives evidence that it was physically dispersed in a convenient manner. This could be considered evidence that a chemical reaction did not occur between filler and resin, which is consistent with the previous results [53].

The mechanical properties of PMMA samples were evaluated through calculating compressive yield strength, modulus of elasticity and hardness. Figure 5 displays the effect of loading content of hybrid SiC/TiO_2 NPs on the compressive yield strength and modulus of elasticity of PMMA denture base material. The results indicate that nanofillers have significant enhance on the strength of the resin. It can be observed that the sample of loading amount of 0.8 wt. % of hybrid SiC/TiO_2 NPs exhibits a maximum improving. So, for this sample, the compressive yield strength and modulus of elasticity were enhanced up to 15.2% and 22.75%, respectively. While the increasing the filler amount above this limit causes an adverse effect on improving the properties of the resin. This demonstrates that low loading of hybrid SiC/TiO_2 NPs has a good bonding reaction. Nevertheless, the hardness value of PMMA samples is displayed in Figure 6. The results of hardness indicate that the pure PMMA have a hardness, D index, of 82.5. It can notice from Fig. 6 that; the hardness significant enhances with increasing of loading amount of the hybrid SiC/TiO_2 NPs. It can be cleared that sample, 2.0 wt.% of hybrid SiC/TiO_2, has a best D index of 88.6 with increasing up to 7.4% comparing with pure sample. Based on the aforementioned results, it is clear that sample, 0.8 wt. %, gives evidence that it has the best mechanical performance.

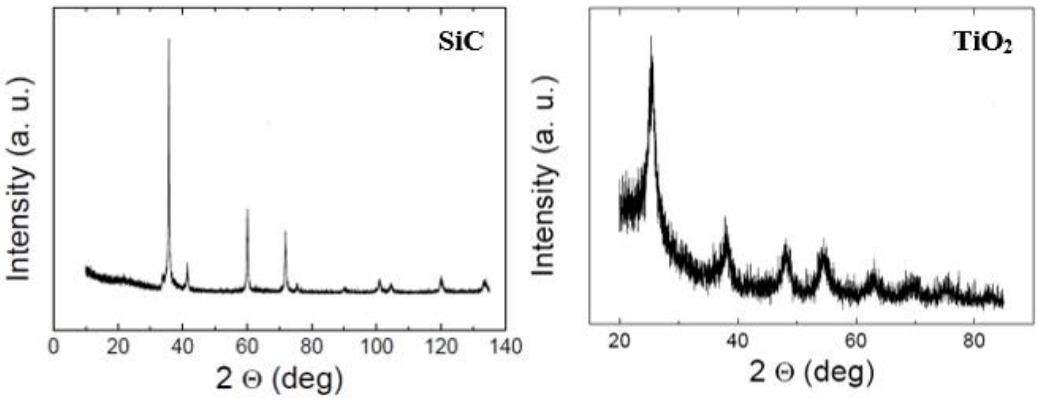

Figure 3. X-ray diffraction pattern for SiC and TiO_2 NPs

Slika 3. Rendgenska difrakcija za SiC i TiO_2 NPs

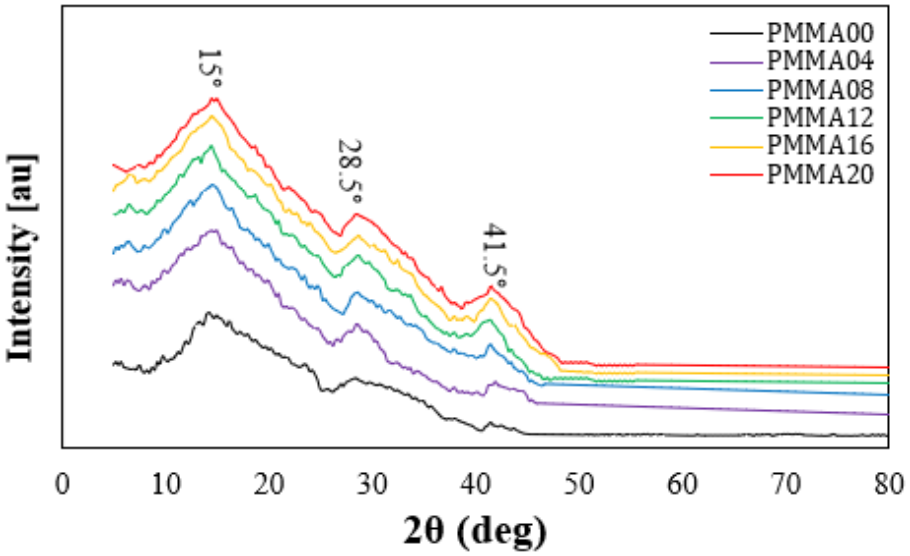

Figure 4. X-ray diffraction pattern for PMMA samples

Slika 4. Difrakcioni rendgenski zraci za uzorke PMMA

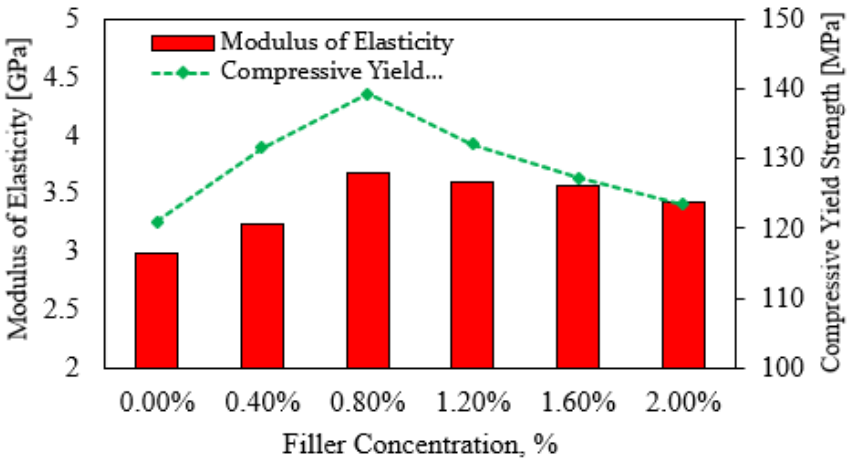

Figure 5. Modulus of elasticity and compressive yield strength of PMMA samples

Slika 5. Modul elastičnosti i granica popuštanja PMMA uzoraka

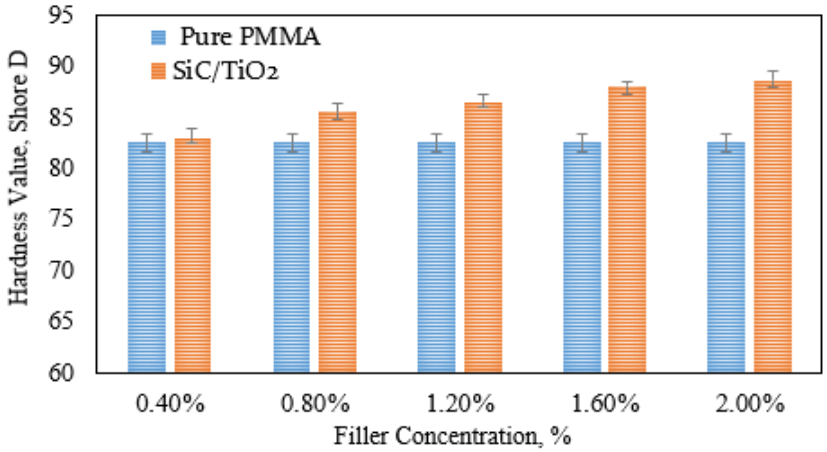

Figure 6. Hardness values of PMMA samples

Slika 6. Vrednosti tvrdoće PMMA uzoraka

The friction coefficient, COF, and wear rate were confirmed to evaluate the tribological performance of PMMA reinforced with SiC/TiO_2 NPs. Figure 7 illustrates the effect of loading content on the COF. It was declared that the maximum COF is archives with the pure PMMA. While PMMA nanocomposite samples stat a good reaction and COF is reduced. This is due to the SiC/TiO_2 NPs have an affect role on reducing COF values. The results reveal that the low filler amount has a higher effect more than the high loading of hybrid filler. Moreover, it may be indicated that sample, 0.8 wt.% of hybrid SiC/TiO_2 NPs, gives a distinctive friction performance. Therefore, it can be verified that COF results of this sample friction are reduced by up to 20% comparing with the pure one. This may be due to form a lubricant thin layer leads the friction between sliding surfaces to reduce. While frictional performance of the samples, with a filler amount above 0.8 wt. %, gave results less than expected. This is likely due to evidence that the bonding between the NPs and the resin is weakened by increasing the amount of filler. Effect of loading content of hybrid filler on the wear rate of PMMA samples was given in Figure 8. The results indicate that increasing of loading content of hybrid NPs leads to reduce the wear rate between the contact surfaces. This can be considered true with loading content up to 0.8 wt. %, because the wear rate begins to rise with increasing filling. It can be concluded that sample with 0.8 wt. % confirms the best result with reduction ratio of 23.4%. This is considered evidence that excessive filling leads to weak internal bonding of the product.

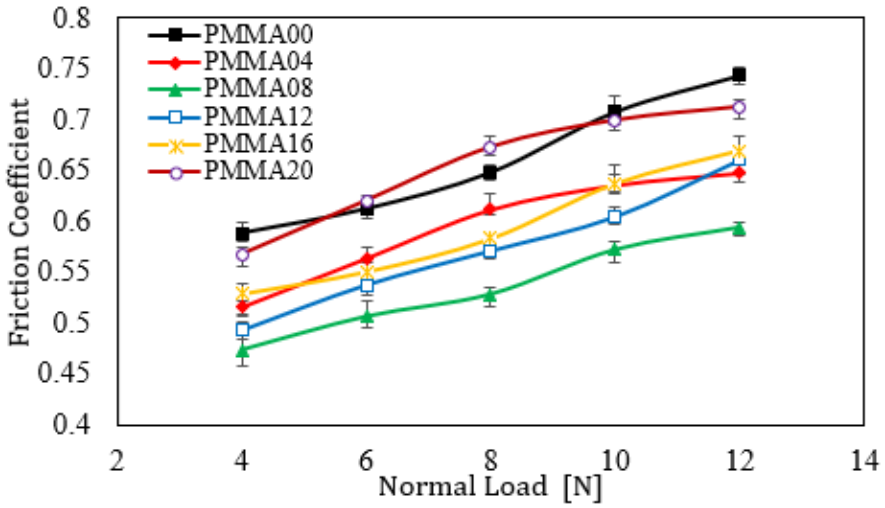

Figure 7. COF of PMMA samples

Slika 7. COF uzoraka PMMA

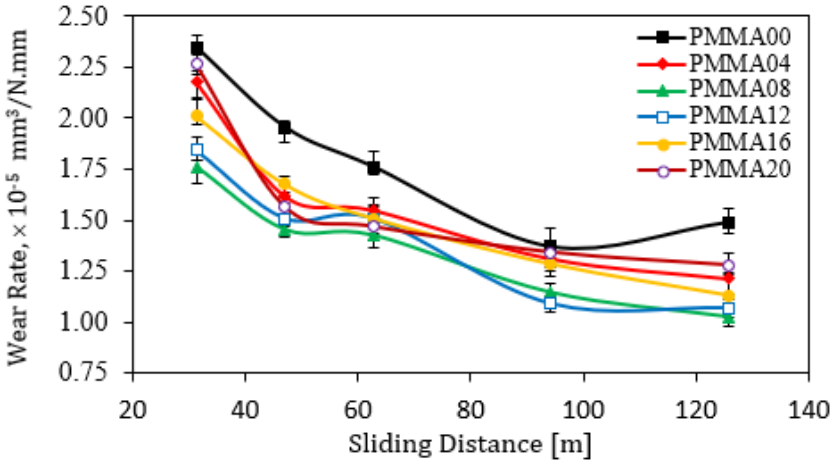

Figure 8. Wear rate of PMMA samples

Slika 8. Stopa habanja PMMA uzoraka

The worn surfaces of PMMA samples were assessed via scanning the frictional surfaces using optical and SEM images. The effect of wear track due to sliding against stainless steel was detailed in Figure 9. The optical topography displays PMMA worn surfaces, which is a sign that the wear is occurred. It can be noticed that contact surface of the tested sample is getting rougher, which is an indication of wear. Which may cause grooves and voids spread on worn surfaces. Moreover, it can be indicated that PMMA sample, with 0.8 wt. % of NPs, exhibits a surface with less groove depth compared to pure sample. In contrast, the surfaces of samples with high loading dispersion of NPS remarkably exhibit the presence of wear tracks and grooves on the worn surface. To verify these results, surfaces examine using SEM analysis. The images obtained from SEM showed a great convergence with the optical images, but with clearer details, as illustrated in Figure 10. It is possible to point out from these SEM images that sample spread on its surface a many of debris, flakes, and delamination. Which gives clear evidence of increasing the weight loss caused by sliding action. Consequently, it can be cleared that the low loading of hybrid SiC/TiO_2 NPs is more suitable for filling this type of resin.

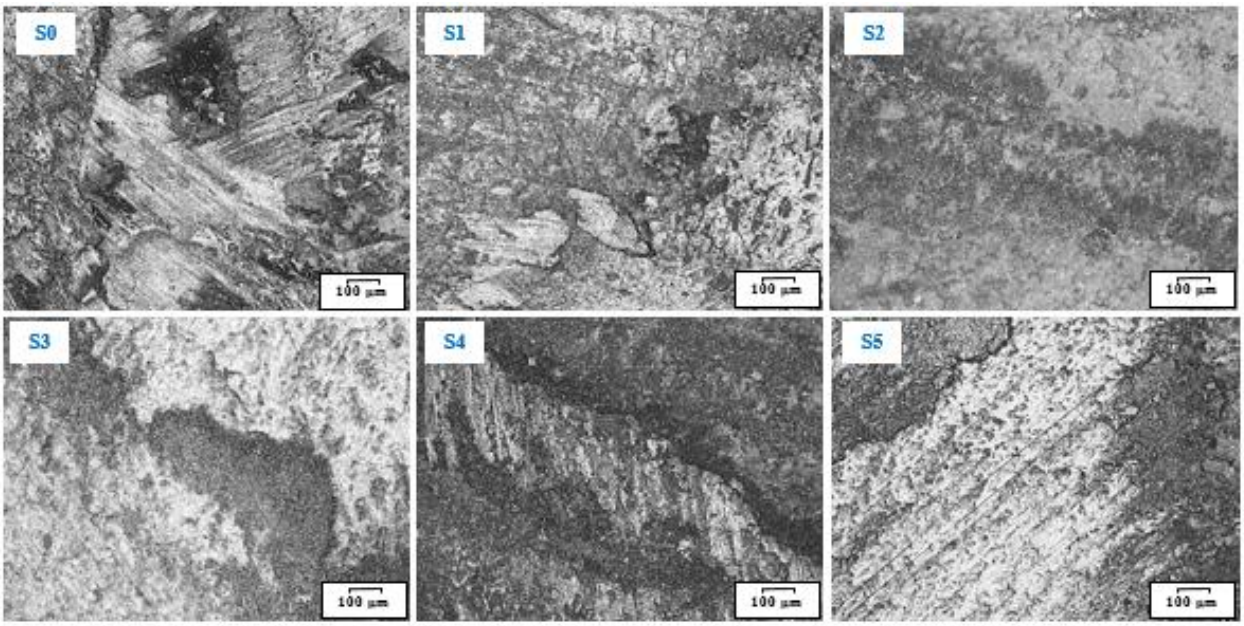

Figure 9. Optical 3D topography of worn surfaces of PMMA samples

Slika 9. Optička 3D topografija istrošenih površina PMMA uzoraka

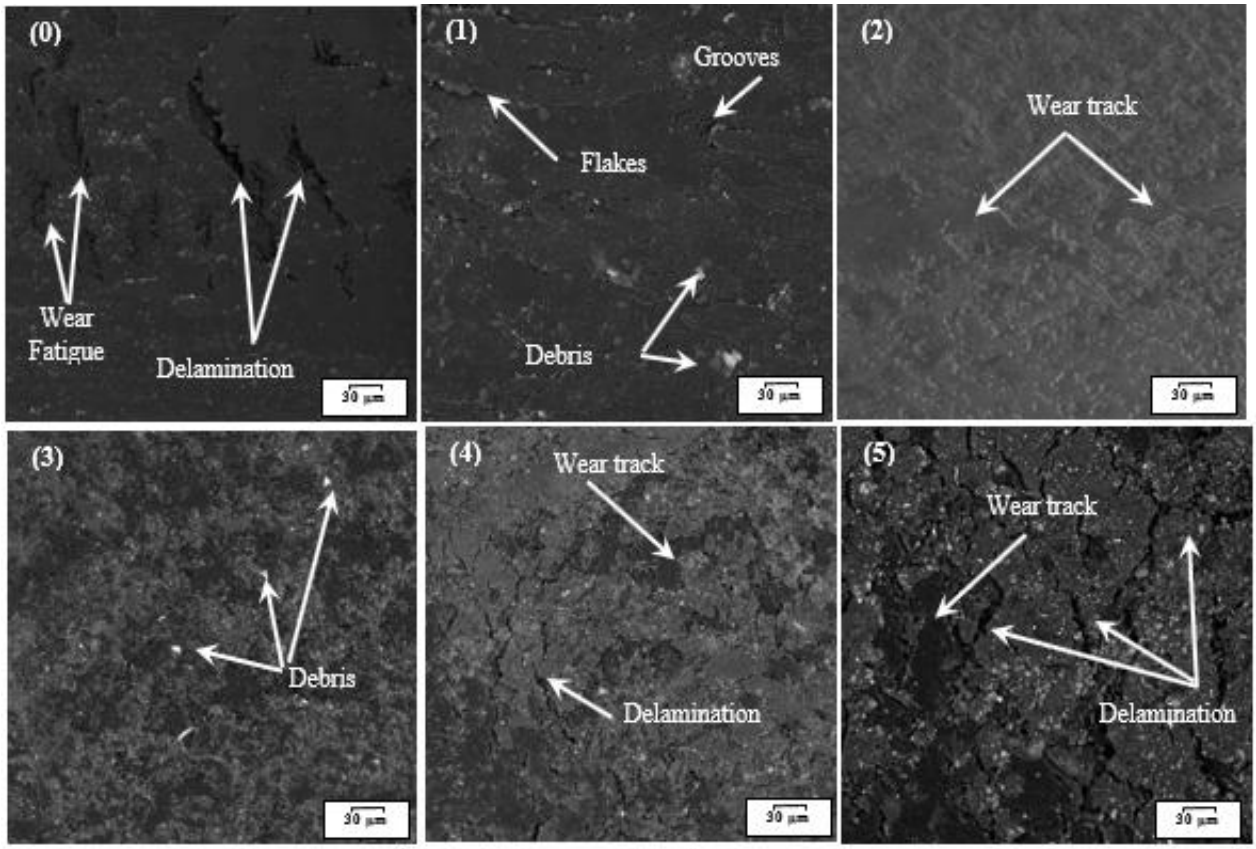

Figure 10. SEM of worn surfaces of PMMA samples

Slika 10. SEM istrošenih površina PMMA uzoraka

4. CONCLUSIONS

This work aims to study the effect of the dispersion loading content of hybrid SiC/TiO_2 NPs on the mechanical and tribological performance. The results indicate that:

- Low loading content of hybrid SiC/TiO_2 NPs have confirmed a good enhancing of tribological and mechanical characteristics of PMMA dental base material comparing with pure PMMA.
- Loading content of 0.8 wt% of SiC/TiO_2 NPs exhibits the best performance comparing with other loading content.
- Loading content of 0.8 wt% of SiC/TiO_2 NPs has a good effect on the compressive yield strength and modulus of elasticity which increased with up to 15.2% and 22.75%, respectively.
- The hardness also improved by 7.4% but the sample with loading content of 2.0 wt%.
- The friction coefficient and wear rate were reduced reached up 20% and 23.4%, respectively for sample with loading content of 0.8 wt%.

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IZVOD

UTICAJ HIBRIDNIH SiC/TiO_2 NANOČESTICA NA TRIBOLOŠKE I MEHANIČKE PERFORMANSE POLIMETILMETAKRILATNOG STOMATOLOŠKOG OSNOVNOG MATERIJALA

Mnoga istraživanja su se bavila PMMA dentalnim osnovnim materijalom i razvojem njegovih svojstava za postizanje dužeg životnog veka implantata i plombi. Sadašnji rad posvećuje pažnju ispitivanju količine opterećenja hibridnih nanočestica, SiC i TiO_2, i saznanju kako to utiče na mehanička i tribološka svojstva. Hibridni NP dispergovani su na PMMA smoli sa količinom punila od 0,4%, 0,8%, 1,2%, 1,6% i 2,0 tež.%, 50/50 između SiC i TiO_2 NPs. Mehanička svojstva su procenjena određivanjem tvrdoće, šor D, čvrstoća na pritisak i modul elastičnosti. Dok su tribološke performanse procenjene ispitivanjem COF-a, stope habanja i skeniranjem istrošenih površina korišćenjem optičkih i SEM slika. Rezultati mogu ukazati na to da PMMA smola pokazuje dobro reakciono vezivanje sa malom količinom hibridnih NPs. Štaviše, visok sadržaj imao je negativan uticaj na mehanička i tribološka svojstva. Nakon toga, sadržaj punjenja od 0,8 tež.% SiC/TiO_2 NPs pokazuje da ima najbolje performanse u poređenju sa čistim PMMA.

Ključne reči: *PMMA, trenje, stopa habanja, mehanička svojstva, SiC nano čestice, TiO_2 nano čestice.*

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