Egyptian Journal of Chemistry
http://ejchem.journals.ekb.eg/

Effect of Low Content of Al_2O_3 Nanoparticles on the Mechanical and Tribological Properties of Polymethyl Methacrylate as a Denture Base Material

Mahmoud Gallab [a,b] *, **Mohamed Taha** [c,d], **Ahmed Rashed** [a], **Ahmed Nabhan** [a]
[a] *Production Engineering and Mechanical Design Department, Minia University, El-Minia 61111, Egypt*
[b] *Micro-Nano Mechanical Science and Engineering Department, Nagoya university, Nagoya 464-8601, Japan*
[c] *Mechanical Engineering Department, College of Engineering and Technology, Arab Academy of Science, Technology, and Maritime Transport, Sadat Road – P.O. Box 11, Aswan, Egypt*
[d] *Department of Materials Engineering and Mechanical Design, Faculty of Energy Engineering, Aswan University, Aswan, 81542, Egypt*

Abstract

In this study, the mechanical and tribological properties of polymethyl methacrylate (PMMA)-based dentures strengthened by a low content of alumina (Al_2O_3) nanoparticles were evaluated. PMMA nanocomposites with different weight fractions (0.2, 0.4, 0.6, 0.8, and 1 wt%) of Al_2O_3 were fabricated, and a sample of pure PMMA was used as a control. Compression, hardness, and wear tests were conducted to investigate the effect of a low Al_2O_3 filler content on the mechanical and tribological properties of PMMA. The addition of Al_2O_3 filler improved the mechanical and tribological characteristics of PMMA. The highest elastic modulus and compressive yield strength (3.56 GPa and 138.21 MPa, respectively) were obtained using the PMMA sample with 0.6 wt% of nanoalumina, which represent an increase of 20.27% and 14.7%, respectively, compared with those of the pure PMMA. The fracture toughness and hardness were improved with the increase in the filler content until it reached 0.8 and 1.00 wt%, respectively. The tribological characteristics of PMMA also improved with the addition of Al_2O_3 nanoparticles. The least coefficient of friction (COF) and wear rate were obtained at a filler content of 0.6 wt%. Therefore, the addition of low amounts of Al_2O_3 to PMMA-based resins can improve their mechanical and tribological properties such as hardness, elastic modulus, fracture toughness, COF, and wear resistance.

Keywords: PMMA composites; Al_2O_3; Mechanical properties; Friction; Wear rate

1. Introduction

Polymethyl methacrylate (PMMA), which has a $(C_5O_2H_8)_n$ chemical formula, is a synthetic acrylic resin based on a methyl methacrylate monomer. Figure 1 shows the chemical structure of the repeat unit of PMMA. PMMA has several advantages such as its superior biocompatibility, exceptional optical clarity, ease of processing to high aesthetic degrees, and low cost [1–3]. Therefore, it is widely used in various practical applications in different fields. Ali et al. presented a comprehensive review of the different properties of PMMA and its applications [4]. In dentistry, PMMA is considered one of the most common materials for denture applications because of its outstanding properties [1]. However, PMMA has some drawbacks regarding its tribological and mechanical properties such as its insufficient wear resistance, hardness, and mechanical strength. Therefore, pure PMMA is not appropriate as a denture base. Thus, PMMA should be enhanced to achieve suitable mechanical characteristics.

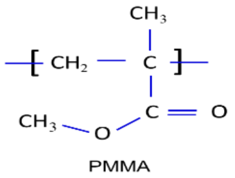

Fig. 1. Chemical structures of poly(methyl methacrylate) polymer repeat unit

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*Corresponding author e-mail: mahmoud@biorobotics.mech.nagoya-u.ac.jp.; (Mahmoud Gallab).

Receive Date: 30 October 2021, Revise Date: 19 December 2021, Accept Date: 10 January 2022
DOI: 10.21608/EJCHEM.2022.88597.4786

In recent years, there has been a growing interest in the improvement of the mechanical properties of composites. One of the strategies that have been used to overcome this drawback is nanoparticle incorporation in the polymer matrix [5–8]. The addition of fillers to the polymer matrix is a common approach to improve the mechanical properties of PMMA-based resins. Numerous studies have examined the improvement of physical and mechanical characteristics using nanocomposites [9]. Several reports investigated dental materials reinforced using micrometer- and nanometer-size materials to create a new composite with enhanced physical and mechanical characteristics [10]. Numerous nanomaterials, such as silver nanoparticles [11], silicon dioxide (SiO_2) [12], platinum nanoparticles [13], carbon nanotubes [14], and hybrid nanomaterials [15], were incorporated with PMMA to develop denture bases with improved physical and mechanical properties. For instance, reinforcing PMMA with multiwalled carbon nanotubes (MWCNT) was shown to improve its mechanical and thermal properties. The author also suggested that the mechanical properties can be further enhanced by using different types of MWCNT [16].

Metal oxides can be used as fillers for PMMA reinforcement to improve their physical and mechanical properties as well as the sensation of the patients of hot and cold stimuli. Hence, reinforced dental base resins can offer better food sensation and healthier oral mucosa [17,18]. Several studies were conducted to investigate the impact of metal oxide fillers on the mechanical and tribological properties of PMMA. Previous studies showed that incorporating zirconia (ZrO_2) nanoparticles in PMMA improved its mechanical properties such as compression strength, fracture toughness, and hardness [19–21]. The wear behavior of PMMA reinforced with ZrO_2 was also investigated. The wear rate of the PMMA/ZrO_2 was notably affected by the percentage of the ZrO_2 additive. The tribological properties of PMMA incorporated with silicon dioxide (SiO_2) nanoparticles were studied by Martinez-Perez et al. [22]. The authors reported that the coefficient of friction (COF) and wear rate decreased with the addition of SiO_2 nanoparticles.

Among the multifunctional metal oxide nanoparticles, Al_2O_3 has excellent advantages such as chemical, optical, and electrical properties; biocompatibility; and low cost. Al_2O_3 nanoparticles have a positive effect on acrylic polymer properties. For example, reinforcing PMMA with Al_2O_3 enhanced its mechanical characteristics such as flexural strength, compressive strength, and surface hardness [17,23,24]. The mechanical characteristics of PMMA reinforced with different types of Al_2O_3, such as nanoparticles, electrospun fibers, and whiskers, were also studied [25]. The authors used high loading fractions (more than 2 wt%) of different types of alumina and found that PMMA reinforced with alumina whiskers has better mechanical properties than the other types of Al_2O_3. Sezavar et al. studied the effect of mass fraction of Al_2O_3 nanoparticles on the fracture behavior of PMMA/Al_2O_3 composites [26]. The authors reported that the addition of alumina to PMMA increased the elastic modulus of the composites but did not affect other properties such as the tensile strength and elongation at break. The thermal stability and flammability of PMMA incorporated with alumina were studied, and the authors reported that the thermal stability was significantly enhanced with the addition of alumina [27]. Previous studies investigated the effect of high fractions of Al_2O_3 nanoparticles on different characteristics of PMMA. However, the high loading fraction of the filler in the polymer caused it to aggregate, hindering the improvement of mechanical and tribological properties because of the poor interaction between the matrix and the filler. Hence, several articles studied the effect of using low loadings of Al_2O_3 nanofillers on different properties of the polymer matrix [28,29]. For instance, the effect of the low loading of Al_2O_3 nanoparticles on the mechanical and tribological properties of epoxy nanocomposites was investigated [28]. The authors found that adding 0.2 vol% of alumina enhances the mechanical and tribological properties of the composite.

To the best of our knowledge, there have been very few investigations regarding the effect of the low contents of Al_2O_3 nanoparticles on the mechanical and tribological properties of PMMA-based dentures. Therefore, in the present study, PMMA reinforced with low content of Al_2O_3 nanoparticles was fabricated, and the effect of Al_2O_3 nanoparticles on the mechanical and tribological properties of the composites was studied. PMMA composites were obtained by loading different concentrations of Al_2O_3 into the PMMA matrix. The specimens were evaluated using compression, hardness, and wear tests, and the different properties of the PMMA composites were evaluated as a function of the Al_2O_3 content.

2. Materials and tests

2.1. Materials

PMMA was purchased from Cold Cure, Acrostone Dental & Medical Supplies, Cairo, Egypt. PMMA consists of two components. One component is monomer liquid component which compromise of methyl methacrylate (MMA), hydroquinone, and N, N-dimethyl para toluidine. The other component traditionally called powder (modified PMMA) which contains PMMA, barium Sulphate, and benzoyl peroxide. Figure 2 shows the chemical stages of the PMMA formation by free radical polymerization of the MMA monomer. Al_2O_3 nanoparticles (80% alpha–20% gamma, purity = 99.9%, particles size = 40–50 nm, specific surface area = 35 m2/g, with nearly spherical morphology, bulk density = 0.18 g/ml, and true density = 3890 kg/m3) were obtained from US Research Nanoparticles, Inc.

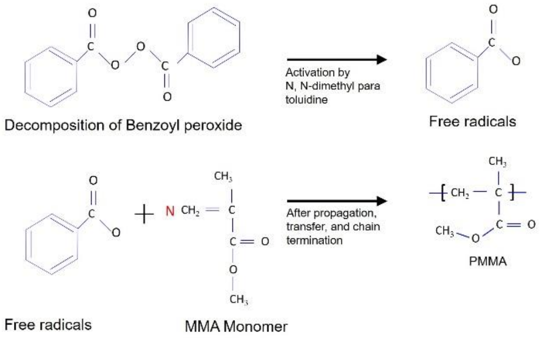

Fig. 2. Chemical stages of PMMA formation

2.2. Specimen preparation

Figure 3a shows the preparation of the PMMA/Al_2O_3 composite. PMMA powder and hardener were blended at a weight ratio of 2:1 as recommended by the manufacture, and Al_2O_3 nanoparticles were dispersed into the PMMA matrix using a rotating mixer at 200 rpm for 10 min. Figure 3b shows a scanning electron microscopy (SEM) image of Al_2O_3 nanoparticles used as the reinforcement for PMMA [30]. Five different PMMA composites (Al_2O_3 wt% = 0.2, 0.4, 0.6, 0.8, and 1 wt%) were fabricated. PMMA samples without any Al_2O_3 nanoparticles were prepared as a control. After an even mixing, the mixture changed into a dough-like mass. The fabricated PMMA matrix was poured into a cylindrical mold and pressed under pressure (10 kp/cm^2) and left to set for 1 h. The inner diameter and height of the cylindrical mold were 8 and 25 mm, respectively. The specimens of PMMA composites were then obtained by demolding them.

2.3. Mechanical properties test

Compression and hardness tests were used to investigate the mechanical properties of the PMMA composites. First, the compression test was conducted at a United High Capacity Smart Universal Hydraulic (DFM-300KN) following the American Society for Testing and Materials (ASTM) standard (D1621). Three test specimens were prepared from the PMMA composites at each filler content to evaluate the compression load-carrying capacity. The stress–strain curves were automatically obtained by the aforementioned machine. The compressive yield strength, modulus of elasticity, and fracture toughness were then determined. Next, the hardness test was conducted to determine the shore hardness (D-index) of the samples on a durometer shore D device with a sharp cone point (SR = 0.1 mm), included angle = 35°, and measuring range of 0–100 ± 0.5 HD. The samples were dwelled for up to 10 s based on the ASTM D2240. Three samples were prepared for each ratio of PMMA composites. For each sample, the hardness value was measured five times along the specimen surface, and then the average value was calculated. The average values and standard errors of all properties were calculated based on the experimental measurements.

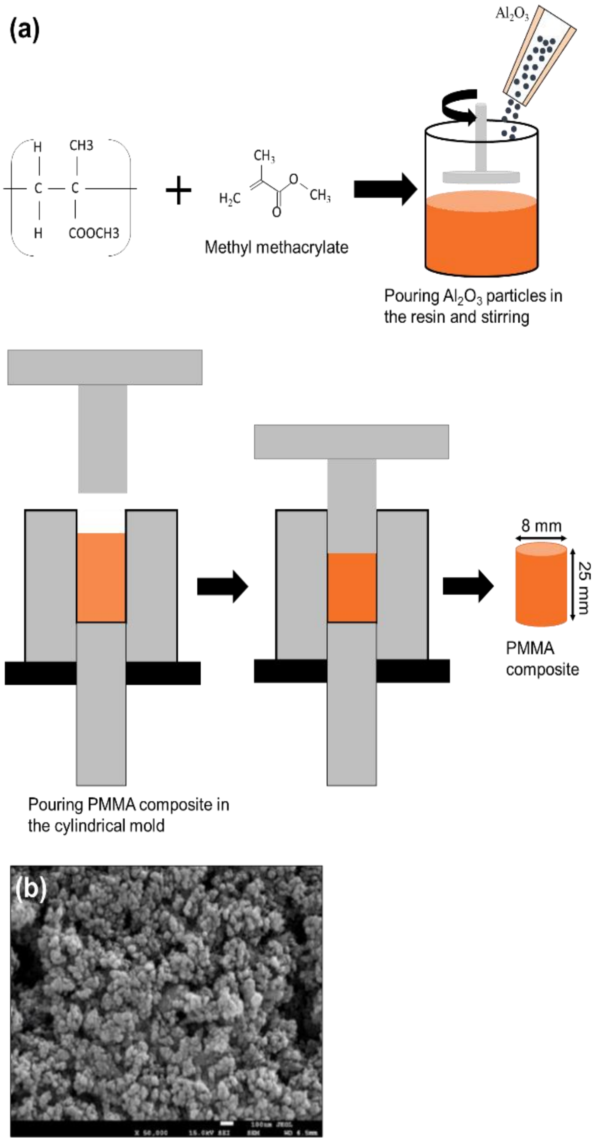

Fig. 3. (a) A schematic diagram of the preparation of PMMA-based nanocomposite specimens. (b) SEM image of Al_2O_3 particles

2.4. Tribological test

The friction and wear properties of PMMA/Al_2O_3 composites were evaluated using a reciprocating pin-on-disk tribometer with a 50 mm stroke (in accordance with ASTM G99-95) under dry contact conditions at an ambient temperature of 29°C and relative humidity of 55% [31]. Figure 4 presents a cad image of the reciprocating pin-on-disk tribometer used in this work. The samples were tested as pins, to which the normal load was applied in a perpendicular direction, and the device had a rectangular disc plate made of stainless steel. The COF was measured under different normal loads (6, 8, and 10 N) at a sliding velocity of 0.4 m/s.

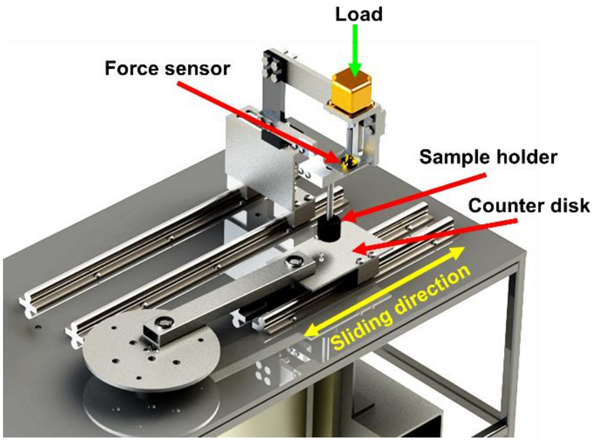

Fig. 4. A cad image for the reciprocating tribometer

Wear was calculated by estimating the weight loss between the initial and final weights of the samples. For each composite composition, the tests were conducted three times under the same conditions and the average reading values were used, and the standard errors were calculated.

2.5. Scanning electron microscopy

The morphological characteristics of the worn surfaces of PMMA composites were observed by SEM (JCM-6000Plus; JEOL, Tokyo, Japan). The samples for this test were spattered with a thin film of platinum to improve their conductivity.

3. Results and discussion

3.1. Mechanical properties

Compression and hardness tests were conducted to investigate the mechanical properties of PMMA composite after the incorporation of a low-weight fraction of alumina nanoparticles in the PMMA matrix. Figs 5, 6, and 7 show the elastic modulus and compressive yield strength, fracture toughness, and hardness of the developed PMMA composites, respectively. The compressive yield strength and modulus of elasticity (Fig. 5) and the fracture toughness (Fig. 6) of pristine PMMA improved when Al_2O_3 nanoparticles were added. This can be attributed to the small particle size and proper distribution of the Al_2O_3 nanoparticles [17]. The highest reinforcement effect of Al_2O_3 on the compressive yield strength and modulus of elasticity was obtained at a 0.6 wt% loading ratio, whereas the maximum fracture toughness was obtained at a 0.8 wt% loading ratio. The compressive yield strength and modulus of elasticity of PMMA/Al_2O_3 increased up to 14.7% and 20.27%, respectively, whereas the fracture toughness increased 10.1% compared with pure PMMA. This increase might be ascribed to the improved stiffness of the PMMA composite achieved from the incorporation of the rigid filler into the PMMA matrix. The addition of Al_2O_3 nanoparticles beyond 0.6 wt% caused a decrease in the compressive yield strength, elastic modulus, and fracture toughness. However, these values were still greater than those obtained for pure PMMA (Figs. 5 and 6). This decrease may be attributed to the tendency of Al_2O_3 particles to aggregate [32], which results in nonuniform dispersion and agglomeration of Al_2O_3 particles, reduces the homogeneity of the mixture, and weakens the matrix when high loadings of Al_2O_3 are used [33,34]. These observations are in agreement with those of Schulze et al. [35]. Schulze et al. suggested that an increase in the filler concentration does not necessarily increase different mechanical properties because a higher filler fraction creates more defects, which weaken the material. Thus, a filler could enhance the mechanical properties of PMMA composites up to a certain amount. Therefore, a PMMA reinforcement of 0.6 wt% Al_2O_3 nanoparticles is the optimum amount that has a significant improvement effect on the mechanical properties.

Figure 7 illustrates the hardness of the prepared PMMA/Al_2O_3 composites at different weight fractions of Al_2O_3. Regardless of the weight fraction of Al_2O_3, all prepared PMMA/Al_2O_3 composites were superior to those of pure PMMA counterparts in terms of hardness. The highest hardness (88.1 on a shore D scale) was obtained at a 1.00 wt% Al_2O_3 loading in PMMA followed by those obtained at 0.80, 0.60, 0.40, and 0.20 wt% Al_2O_3 loadings (86.5, 85.5, 84.2, and 82.3, respectively). These values represent an increase in the hardness by 2.0%, 4.3%, 5.9%, 7.2%, and 9.2%, respectively, compared with pure PMMA (80.7).

Thus, a low loading content of Al_2O_3 has a significant effect on the mechanical properties of PMMA-based denture material. Based on the previous results, a reinforcement of 0.6 wt% Al_2O_3 nanoparticles can significantly improve the mechanical properties of PMMA-based composites.

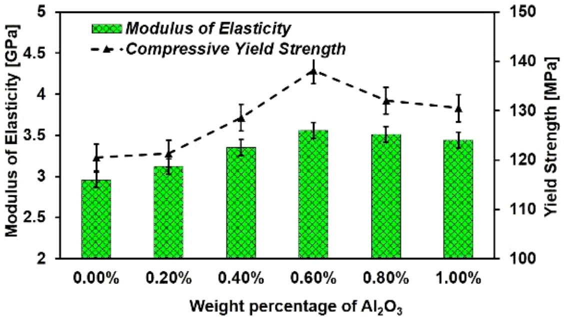

Fig. 5. Compressive yield strength and modulus of elasticity of the PMMA/Al_2O_3 samples

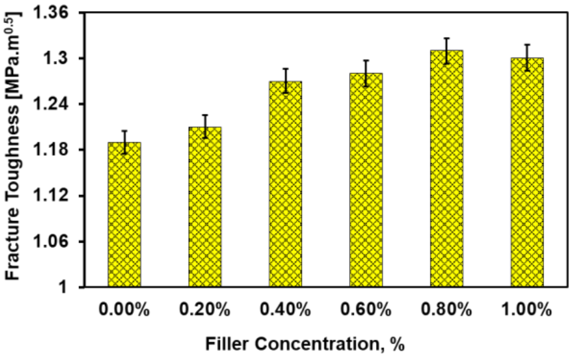

Fig. 6. Fracture toughness of the PMMA/Al_2O_3 samples

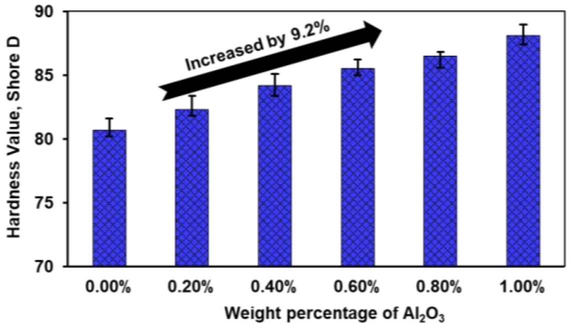

Fig. 7. Hardness of PMMA/Al_2O_3 composites at different weight fractions of the filler

3.2. Tribological properties

PMMA is rarely used as a tribological material because it has a relatively low wear resistance compared with glass [36]. Here, the effect of low loading contents of Al_2O_3 on the tribological properties of PMMA was studied. Thus, the fabricated samples were rubbed against a PMMA disk under different normal loads (6, 8, and 10 N) at a sliding velocity of 0.4 m/s, and the average COF was determined. Figure 8 shows the average COF of PMMA composites as a function of the loads applied during the rubbing of PMMA nanocomposites. Figure 8 shows the dependence of the COF values on the Al_2O_3 content in the PMMA composite and the applied loads. Higher COF was measured at higher applied loads. The COF increased with the increase in the normal load in both neat PMMA and PMMA nanocomposites. This may be attributed to the increase in the normal load that causes a high contact pressure between the sliding faces, which increases the frictional heat and, consequently, the COF [37]. Moreover, the incorporation of hard particles, such as Al_2O_3, was found to decrease the COF. The decline in COF might also be attributed to the hindering effect of Al_2O_3 on the delamination and adhesion of the PMMA matrix. However, the COF of the PMMA composites increased at filler contents higher than 0.6 wt%. This may be ascribed to the agglomeration of Al_2O_3 because of its high surface energy, which causes an abrasive impact during the test as the Al_2O_3 nanoparticles had an abradant effect on the PMMA surface, which increased the COF. The shape and abrasiveness of the reinforcement particles also affected the value of COF [38].

Figure 9 shows the results of the wear measurements of PMMA composites. The weight loss and wear rate due to the applied load were measured. The wear rate of PMMA decreased by 8.44% at an Al_2O_3 nanoparticle content of 0.2 wt%. The maximum reduction in the wear rate was 26.3% at a filler content of 0.6 wt%. The wear resistance improved because PMMA was less exposed to the abrasion plate. Instead, the alumina nanoparticles were the ones exposed to the abrasion disc, which decreased the contact area between the disc and the PMMA composite. Additionally, the increase in the strength and hardness of the PMMA composite enhances the wear resistance because of the increase in the impedance against indenter penetration. However, the addition of Al_2O_3 beyond 0.6 wt% caused an increase in the wear rate. This can be attributed to the increase in Al_2O_3 dispersion, which increased the agglomeration of Al_2O_3. Some of the agglomerated particles were abraded upon touching the abrasion disc, and a three-body abrasion occurred, causing an increase in the wear [39]. Thus, the addition of Al_2O_3 nanoparticles to pure PMMA improves its wear resistance.

Figure 10a–c illustrates optical images of some selected worn pin surfaces of pure PMMA and PMMA reinforced with 0.4, 0.6, and 1 wt% Al_2O_3. The worn surfaces of the selected samples clearly reflect the Al_2O_3 loading-dependent wear behavior. Figure 10a indicates that the worn surface had deep grooves. Pure PMMA exhibited the highest COF (Fig. 8), reflecting the high contact between the surfaces, which resulted in the observed deep grooves (Fig. 10a). Figure 10b and c shows the worn surfaces of PMMA with 0.4 and 0.6 wt% of Al_2O_3 nanoparticles, respectively. The figure indicates that the smoothness of the worn surface increased with the addition of Al_2O_3 nanoparticles. This increase in the worn-surface smoothness can be attributed to the load withstand capacity of the Al_2O_3 particles as well as the large hardness and strength of PMMA composites, which caused an increase in the wear resistance of the PMMA composites during the wear tests. Consequently, the surface morphology changed from a surface with deep grooves (Fig. 10a) to a smooth surface with small scratches (Fig. 10b and c). Regardless of the abrasive properties of alumina nanoparticles, their addition to PMMA improves its wear resistance [32,40,41]. However, the wear rate of PMMA composites increased at a filler content larger than 0.6 wt% (Fig. 9). The optical image of the worn surface of a PMMA composite with a filler content of 1.00 wt% shows a deep groove (Fig. 10d). As mentioned, the increase in the filler resulted in its agglomeration, increasing the wear [39]. Figure 11 presents SEM images of worn surfaces of some composites after abrasion. Figure 11a shows debris, voids, and areas of delamination over the entire surface of the pure-PMMA sample. The peeling and wear debris on the contact zone are evidence of the high wear rate and COF of the pure sample. Figure 11b and c shows the SEM images of the worn surface of a PMMA filled with 0.2 and 0.6 wt% of Al_2O_3 nanoparticles, respectively. The addition of Al_2O_3 nanoparticles reduced the delamination areas, which can be attributed to the high hardness of the nanoparticles. Moreover, the rolling effect of Al_2O_3 nanoparticles reduced the frictional contact forces. Therefore, increasing the content of Al_2O_3 nanoparticles resulted in a low wear rate. However, the SEM image of PMMA incorporated with 1.00 wt% shows a higher wear rate than PMMA incorporated with 0.60 wt% (Fig. 11d). As mentioned earlier, this can be attributed to the increase in the dispersion of Al_2O_3, leading to an aggregation of Al_2O_3 and an increase in the wear rate. Nevertheless, the wear rate of PMMA/Al_2O_3 is still lower than that of pure PMMA. Therefore, increasing the content of Al_2O_3 nanoparticles has a positive effect on the tribological properties of PMMA.

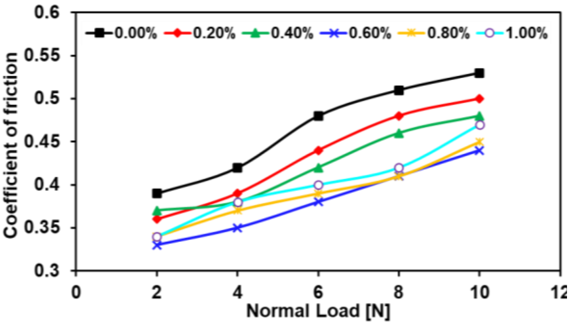

Fig. 8. The coefficient of friction of PMMA/Al_2O_3 samples at different normal loads

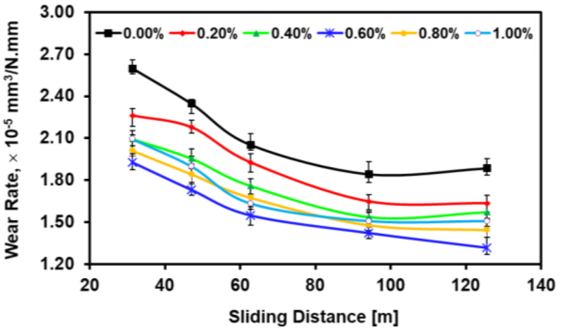

Fig. 9. The wear rate of PMMA/Al_2O_3 samples at a normal load of 10 N with different sliding distances

4. Conclusions

In this study, PMMA nanocomposites reinforced with Al_2O_3 nanoparticles were prepared to investigate the effect of low loadings of Al_2O_3 on the mechanical and tribological properties of the composites. The results indicated that the elastic modulus, hardness, and fracture toughness, were improved after the reinforcement with Al_2O_3 nanoparticles compared with pristine PMMA. The optimum content of the filler was found to be 0.6 wt%. The PMMA composites had the highest wear resistance and least coefficient of friction when the Al_2O_3 nanoparticles content was 0.6 wt%. Briefly, the mechanical and tribological properties of PMMA were improved with low loading of alumina nanoparticles.

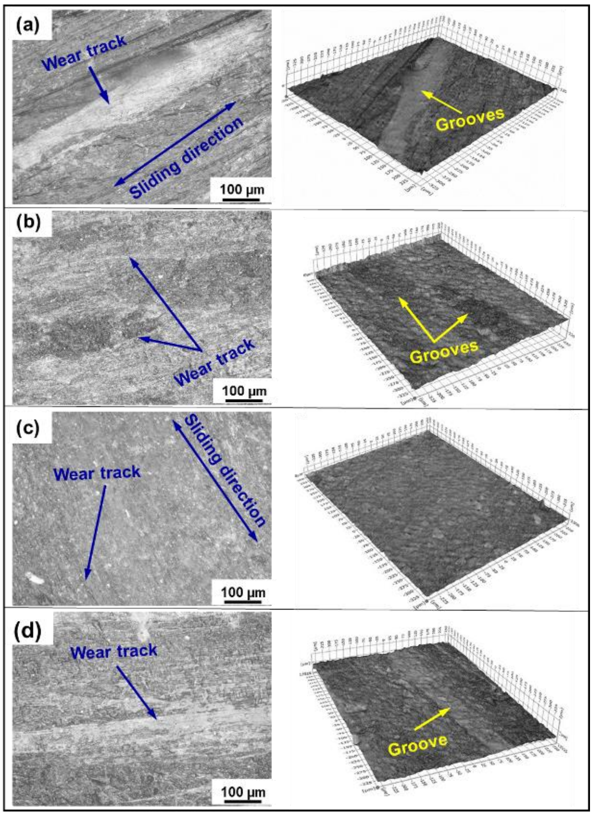

Fig. 10. Optical images and 3D topography showing the surface roughness of worn surfaces of (a) pure PMMA, (b) PMMA + 0.20 wt% AL_2O_3, (c) PMMA + 0.60 wt% AL_2O_3, and (d) PMMA + 1.00 wt% AL_2O_3 tested under a load of 10 N

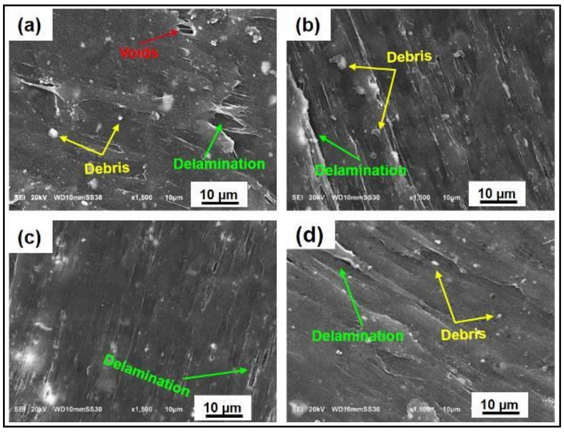

Fig. 11. SEM images of worn surfaces of (a) pure PMMA, (b) PMMA + 0.20 wt% AL_2O_3, (c) PMMA + 0.60 wt% AL_2O_3, and (d) PMMA + 1.00 wt% AL_2O_3 tested under a load of 10 N

5. Conflicts of interest

There are no conflicts to declare.

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