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FRICTION AND WEAR OF POLYMETHYL METHACRYLATE REINFORCED BY SILICA AND ZIRCONIA NANOPARTICLES AS DENTAL MATERIALS

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ABSTRACT
In this study, unfilled PMMA and two groups of specimen composites of polymethyl methacrylate (PMMA) were investigated. The first group was reinforced with different contents of SiO_2 nanoparticles ranging from 2.5 wt. % to 15 wt. % in order to obtain the optimum composition of the mixture that maintains the excellent tribological properties over that of the unfilled PMMA. The second group was prepared by adding zirconia (ZrO_2) nanoparticles of content ranging from 2.5 % to 7.5 % to the composite containing 7.5 wt. % SiO_2 nanoparticles.

The effect of reinforcing PMMA with silica and zirconia nanoparticles on the wear and friction was investigated under dry sliding conditions, constant sliding velocity of 2 m/s and different applied normal load values, 5, 10, 15, 20 and25 N. The time of experiment was 600s. Based on the experiments carried out in the present work, it was found that wear resistance and friction coefficient of the composites were significantly influenced by SiO_2 and ZrO_2 nanoparticles contents. The lowest wear value in (PMMA/SiO_2 nanoparticles) composite was obtained for 7.5 wt. % of silica nanoparticles, while for PMMA/SiO_2 and ZrO_2 nanoparticles was reported for 7.5 wt. % of silica and 5 wt. % of zirconia nanoparticles.

KEYWORDS
Polymethyl methacrylate, silica, zirconia, nanoparticles, wear, friction coefficient, restorative dental materials.

INTRODUCTION
Revolutions in the field of nanotechnology and nanomaterials in various domains including healthcare have made great attention towards the use of nanoparticles in various specialties of dentistry. Nanoparticles (NPs) are insoluble particles smaller than 100 nm in size and the set of technologies that enables manipulation of these particles on

an atomic, molecular and supra molecular scale is termed as 'Nanotechnology', [1]. Nanoparticles have many widely known intrinsic characteristics relating to their composition, size, and shape plus their ability to enhance the existing properties of polymers and even create new ones, [2]. Although dental implants are increasingly used in the treatment of edentulous patients, in many cases a conventional complete denture is still the treatment of choice for medical and financial reasons, [3]. For edentulous patients, the longevity of dentures depends substantially on the wear resistance of denture teeth. As this property may affect the ability of denture teeth to preserve a stable occlusion overtime.

The leading material for dentures nowadays is polymethyl methacrylate (PMMA). It possesses poor mechanical and physical properties when used alone, where it is easilybroken intoparts when a patient applies high mastication force. Its advantages are related to its biocompatibility and esthetics, whereas its drawbacks are insufficient ductility and strength, which leaves opportunities for further improvement, [4, 5]. Several studies have been conducted with the goal of enhancing the properties of PMMA by using different curing methods and/or incorporating fillers in its composition, [6, 7]. Addition of fillers and fibers to PMMA is a commonly used method to improve both its physical and mechanical properties.

Wear resistance of denture teeth has been considered as one of the most important requirements for oral rehabilitation of edentulous patients with removable dentures, in order to maintain a stable occlusal support over time, [8]. Wear of the occlusal surfaces may result in insufficient posterior tooth support, [9], loss of chewing efficiency, [10] and nonfunctional activities, [11]. Initially, denture teeth were made of ceramic material. With the advent of poly methyl methacrylate (PMMA), a new material was introduced for the fabrication of denture teeth, [12]. Denture teeth are currently made of either methacrylate based resins (acrylic resin) or ceramics, although resin teeth have almost eliminated ceramic teeth from the market, [9], due to a number of advantages such as the chemical bond to denture base, [13], lower susceptibility to fracture, [14] and decrease of clicking, [15]. Nonetheless, the wear resistance of acrylic resin teeth has been questioned for being lower than that of ceramic teeth. Manufacturers have then tried to develop acrylic resins designed to offer improved wear resistance for resin denture teeth, [16].

Composite material is a heterogeneous combination of two or more materials, differing in form or composition. The combination results in a material that exploits the (mechanical, electrical, thermal and chemical) properties of individual constituents and maximizes specific performance properties of the composite, [17]. Composite resin denture teeth were developed in the 1980s as an effort to achieve greater wear resistance and bond strength to denture bases, [18]. Tooth materials made with micro particle inorganic fillers immersed in a BIS-GMA (bisphenol a glycidyl methacrylate) matrix, or Nano metric inorganic fillers immersed in a PMMA matrix, [19], have been used for fabrication of composite denture teeth. It has been reported that composite denture teeth show a higher wear resistance than acrylic resin denture teeth. Nano oxide as SiO_2, TiO_2, and ZrO_2 are characterized by their small size, large specific surface area, active function, and strong interfacial interaction with the organic polymer, [20]. Recently,

nanometer inorganic compounds, such as titanium dioxide (TiO_2), zinc oxide (ZnO), silica (SiO_2), aluminum dioxide (Al_2O_3), silicon nitride (SiN_2), and so on, were tried as the fillers of fabric composites and polymers to improve the tribological properties, [21].

The present work investigates the effect of reinforcing PMMA with SiO_2 and ZrO_2 nanoparticles on the wear and friction coefficient.

EXPERIMENTAL

Tribological tests were conducted using pin-on-disc wear tester, general view of the test rig is shown in Fig.1. It consists of a horizontal rotary steel disc driven by a motor of variable speed. The specimen is held in the specimen holder that is fixed to the loading lever through load cell connected to Arduino Uno board to computer. Through the deflection of the load cell, the friction force can be measured and recorded every second on the computer then friction coefficient was determined. The counterface in the form of a carbon steel disc was fastened to the rotating disc. Its surface roughness (Ra) was about 3.2 µm, 2720 N/mm² hardness and 200 mm diameter. Wear tests were carried out under constant sliding velocity of 2m/s and different applied load values 5, 10, 15, 20 and25 N. Every experiment lasted for 600s. Wear was measured by the difference between the weights of the specimens before and after the test using a digital electronic balance of + 0.1 mg accuracy.

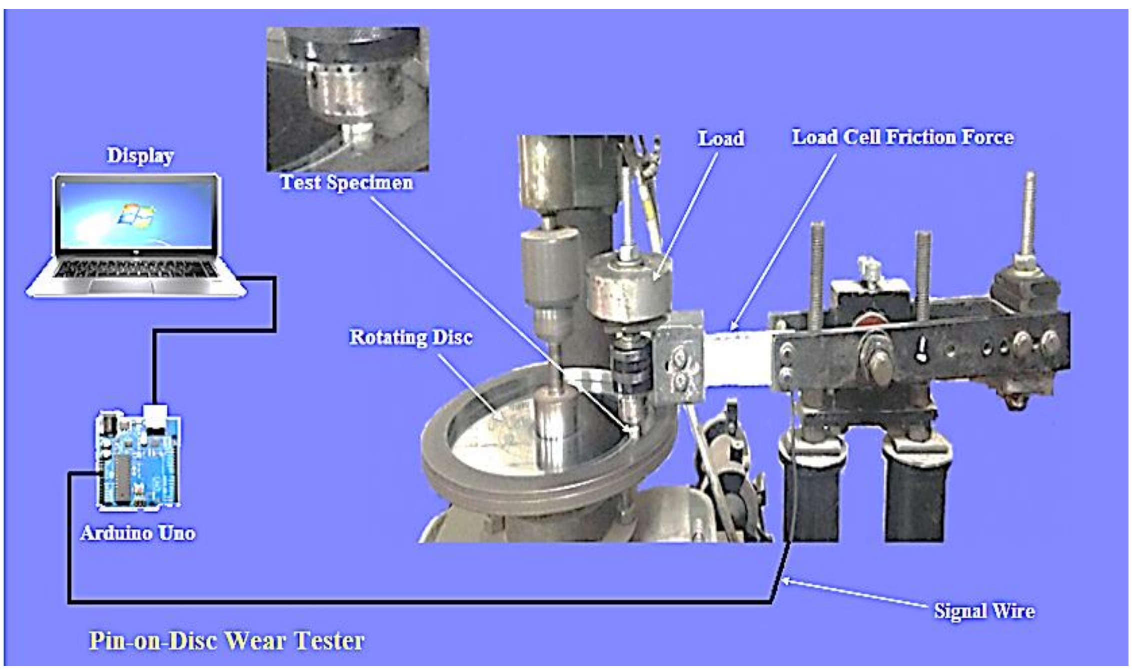


Fig. 1 General view for the pin-on-disc wear tester.

In this study, heat cured PMMA was used as polymer matrix. Unfilled PMMA and two groups of specimen composites were investigated. The first group was reinforced with 2.5 wt. %, 5 wt. %, 7.5 wt. %, 10 wt. %, 12.5 wt. % and 15 wt. %of SiO_2 nanoparticles. The optimum composition that gives the excellent tribological properties was determined. To more improve of the wear resistance, the second group was accomplished by adding different weight percentages of zirconia (ZrO_2) nanoparticles,

2.5 wt. %, 5 wt. % and 7.5 wt. %. The optimum weight percentage to get the minimum wear value for (PMMA/SiO_2 and ZrO_2 nanoparticles) composite was determined.

The diameters of silica and zirconia nanoparticles were 40 nm and 35 nm respectively. Test specimens have been molded in form of cylindrical tube of 9 mm diameter and 30 mm length. Then they have been heat cured with conventional water bath method for 2 hours at 100 ˚C.

RESULTS AND DISSCUSIONS

The influence of the normal load on the friction coefficient during the test for heat cured unfilled PMMA is shown in Fig. 2. It is obviously clear that, friction coefficient increases up to maximum at 15 N, then decreases with further load increase. It can be seen that, the highest friction increase was observed at the beginning of the experiment due to polymer transfer to the steel disc. Silica nanoparticles have a significant role in nanotechnology, due to its size, surface area, biocompatibility, low toxicity, low density and adsorption capacity, [22].

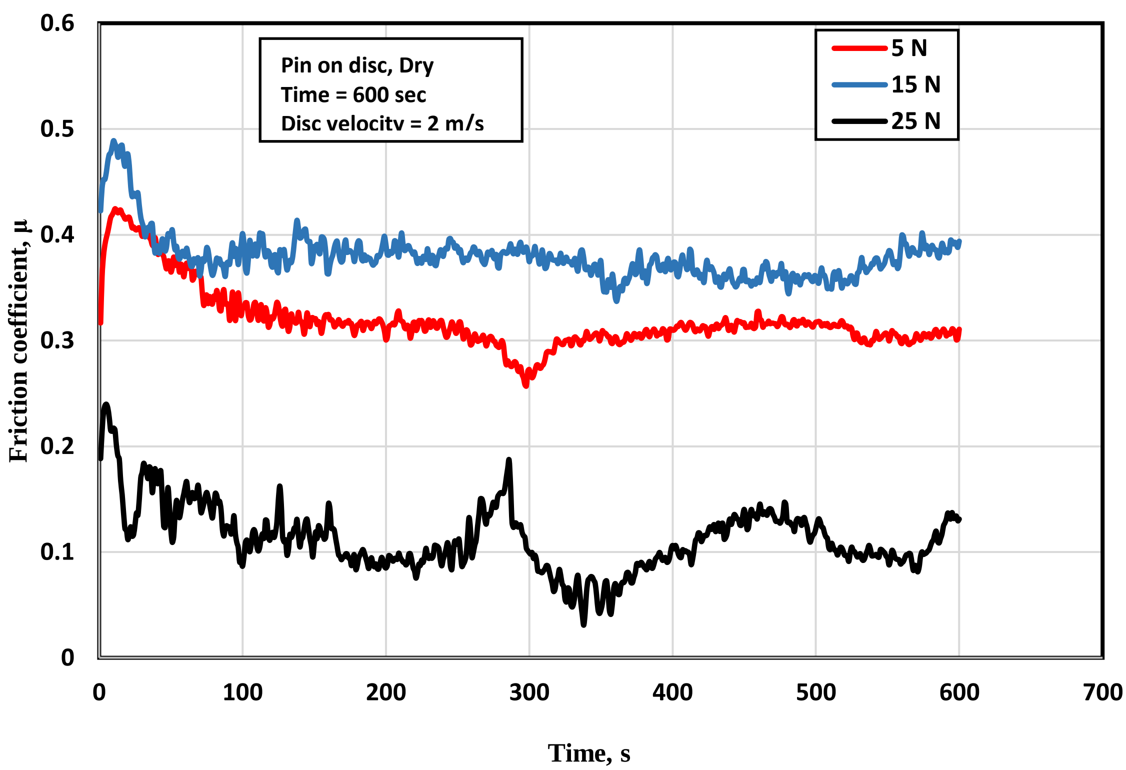


Fig. 2 The influence of the normal force on the friction coefficient for heat cured unfilled PMMA.

Figures 3 and 4 illustrate the effect of applying normal force on the friction coefficient during the test for heat cured PMMA composite reinforced with 7.5 and 15 wt. % of silica nanoparticles respectively. It is clear that, the trends have the same phenomena like that shown in Fig. 2, but it can be seen that, the minimum friction coefficient values

were observed for 7.5 wt. %, 15 wt. % and unfilled PMMA respectively at the same applied normal load. The friction increase may be from the polymer transfer from the specimen to the counterface then after a while, polymer will transfer back to the specimen, so that friction will be between the polymer of the specimen and the polymer on the counterface that will lead to friction increase. It can be seen that, Fig. 4, friction fluctuations increased due to the increase of SiO_2 content, where SiO_2 nanoparticles abraded the polymer and caused significant increase in friction fluctuations.

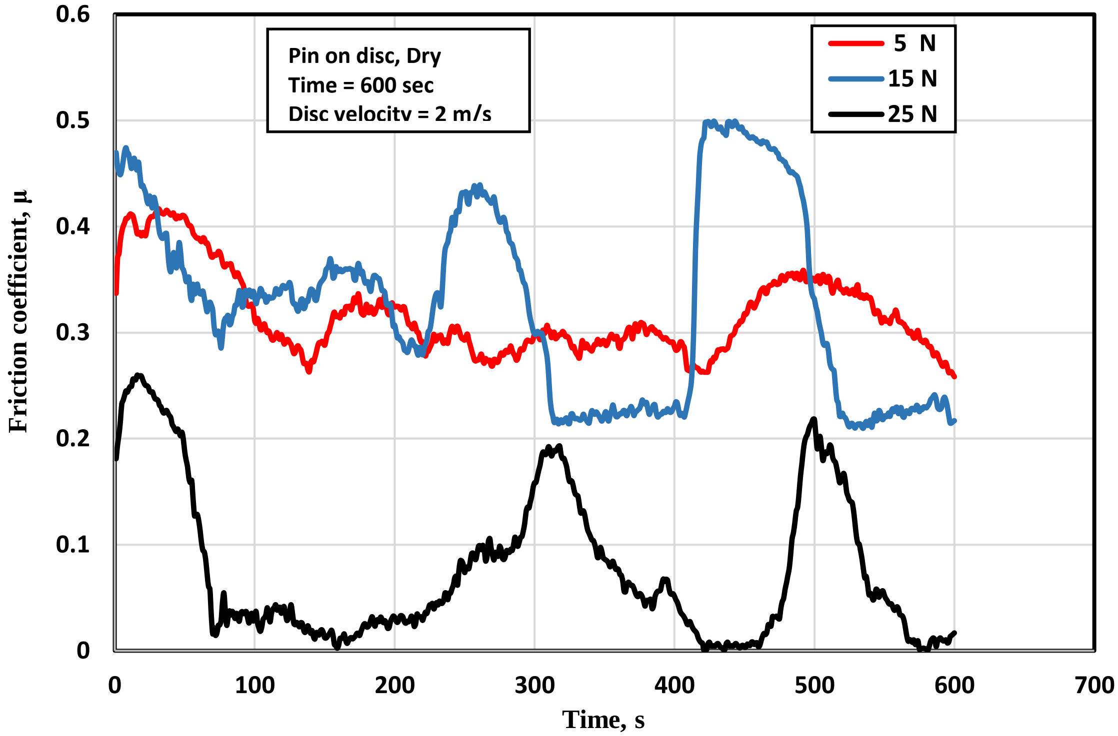


Fig. 3 The influence of the normal force on the friction coefficient for heat cured PMMA reinforced with 7.5 wt. % of SiO_2 nanoparticles.

The relation between the normal load and the wear of heat cured unfilled PMMA and PMMA reinforced with different weight percentages of silica nanoparticles is illustrated in Fig. 5. It is clear that, the minimum wear value was observed for PMMA composite reinforced with 7.5 wt. % of silica nanoparticles while the maximum wear values were observed for unfilled PMMA and PMMA reinforced with 15 wt. % of silica nanoparticles. It is obviously seen that, for unfilled PMMA and (PMMA/SiO_2 nanoparticles) composites, wear increases with the increment of the applied normal load.

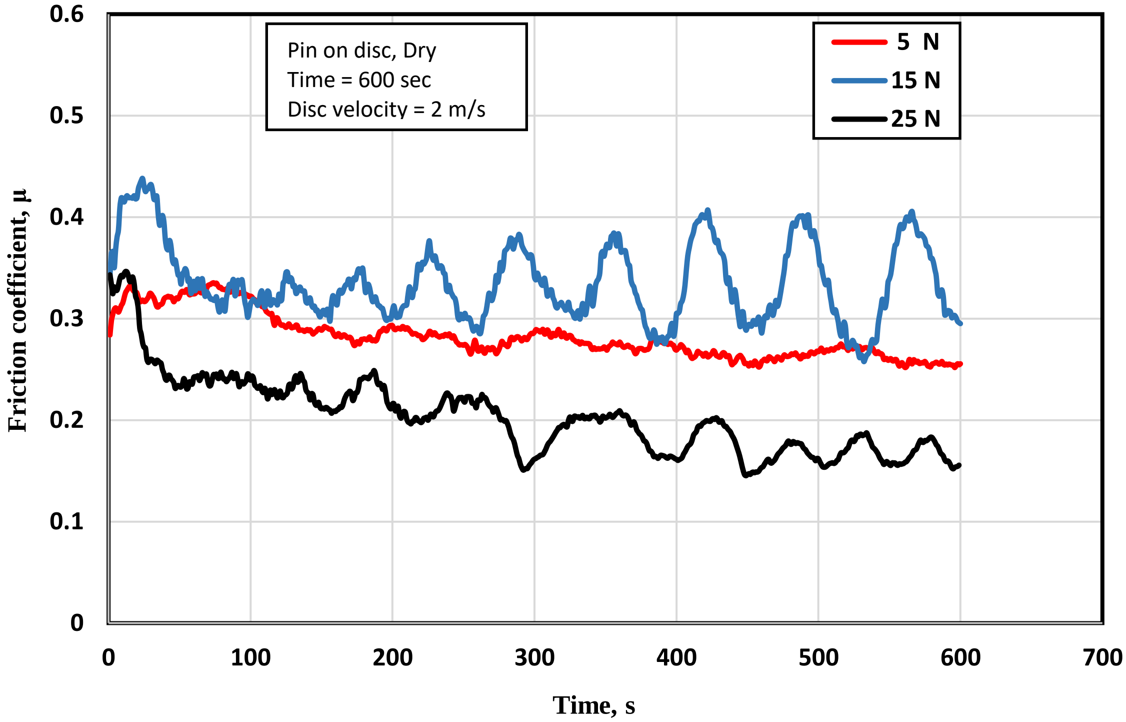


Fig. 4 The influence of the normal force on the friction coefficient for heat cured PMMA reinforced with 15 wt. % of SiO_2 nanoparticles.

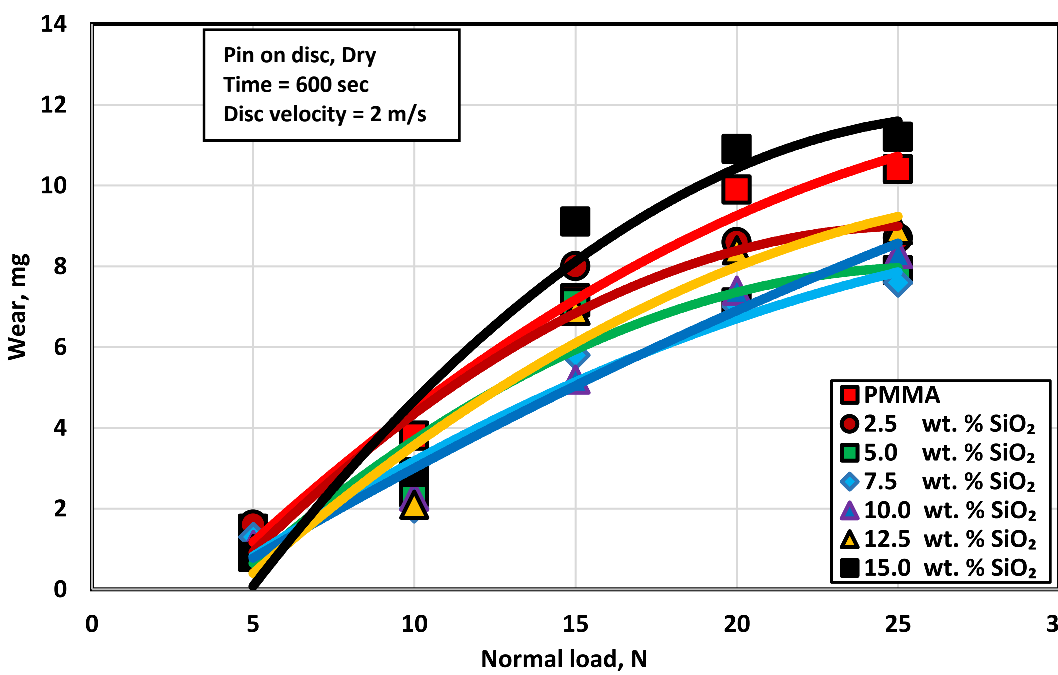


Fig. 5 The relation between the normal load and the wear for heat cured PMMA reinforced with different weight percentage of SiO_2 nanoparticles.

Figure 6 shows the relation between the normal load and the friction coefficient of heat cured unfilled PMMA and PMMA reinforced with different weight percentages of silica nanoparticles. It is clearly shown that, the friction coefficient depends on the applied normal load. It can also be seen that, the normal load at 25 N caused significant friction decrease may be due to the heating effect that decreases the shear strength of PMMA matrix.

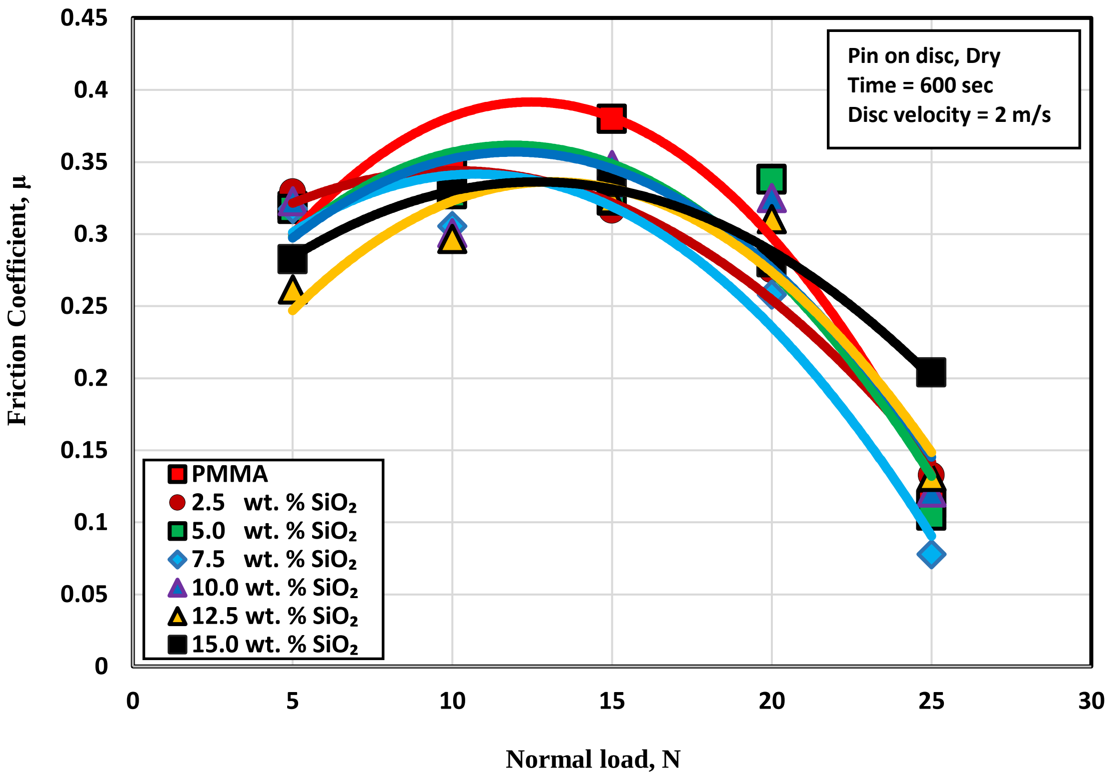


Fig. 6 The relation between the normal load and the average friction coefficient for heat cured PMMA reinforced with different wt. % of SiO_2 nanoparticles.

The influence of the normal load on the friction coefficient during the test for heat cured PMMA composite reinforced with (7.5 wt. % SiO_2 and 5 wt. % ZrO_2) nanoparticles is shown in Fig.7. ZrO_2 possesses excellent properties like; high strength, high fracture toughness, excellent wear resistance, high hardness, and excellent chemical resistance, [23].Good adhesion and dispersion homogeneity of nano-ZrO_2 with the resin matrix effectively improve the properties of the polymer/nanoparticles composite, [24]. During the test, relatively high friction fluctuations were observed. This behavior may be due to the transfer of PMMA into steel disc and back to composite surface.

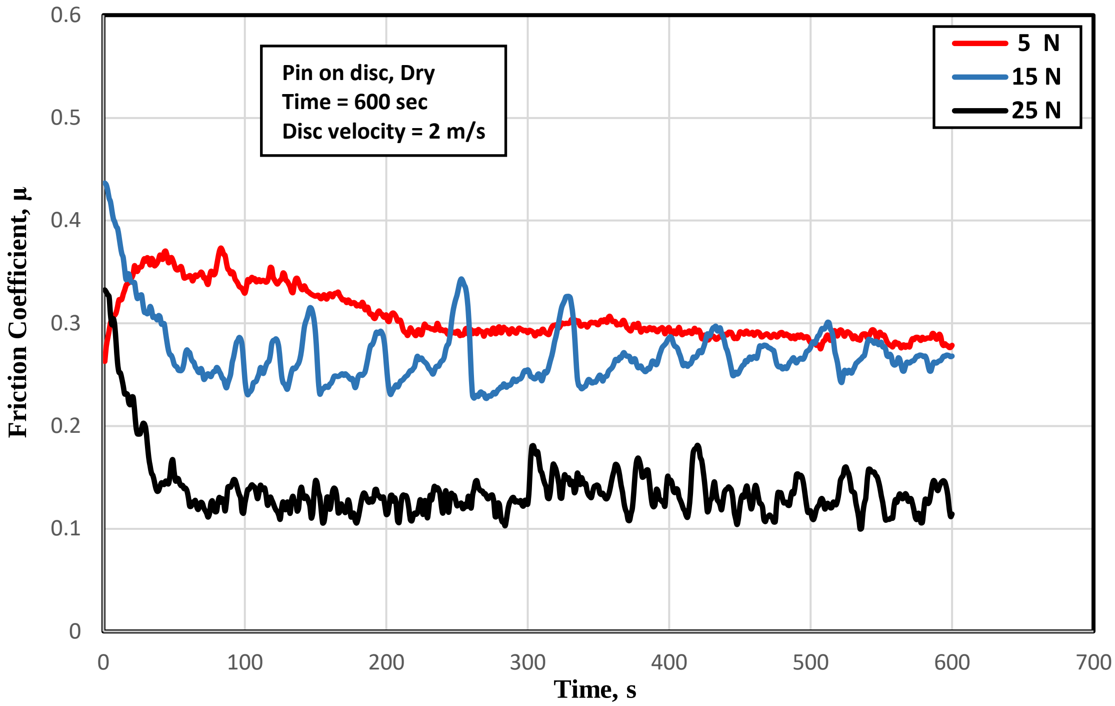


Fig. 7 The influence of the normal force on the friction coefficient for heat cured PMMA reinforced with (7.5 wt. % SiO_2 and 5 wt. % ZrO_2) nanoparticles.

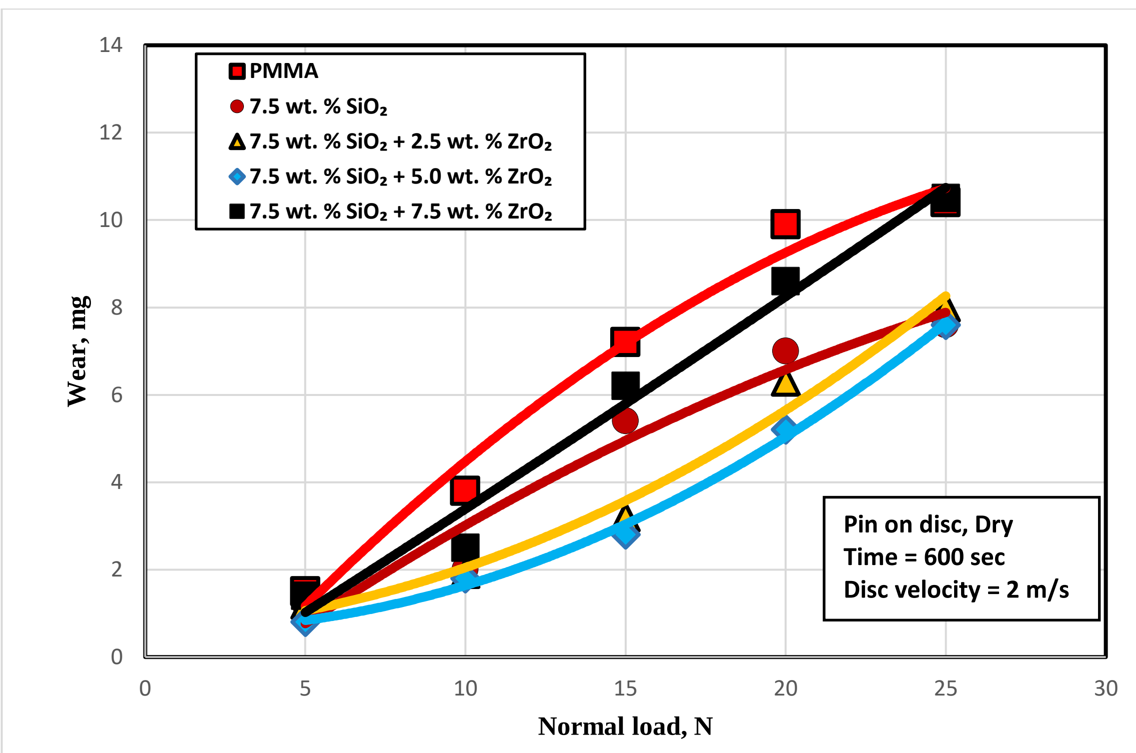


Fig. 8 The relation between the normal load and the wear for heat cured unfilled PMMA and (PMMA /7.5 wt. % SiO_2 nanoparticles) composite reinforced with different content of ZrO_2 nanoparticles.

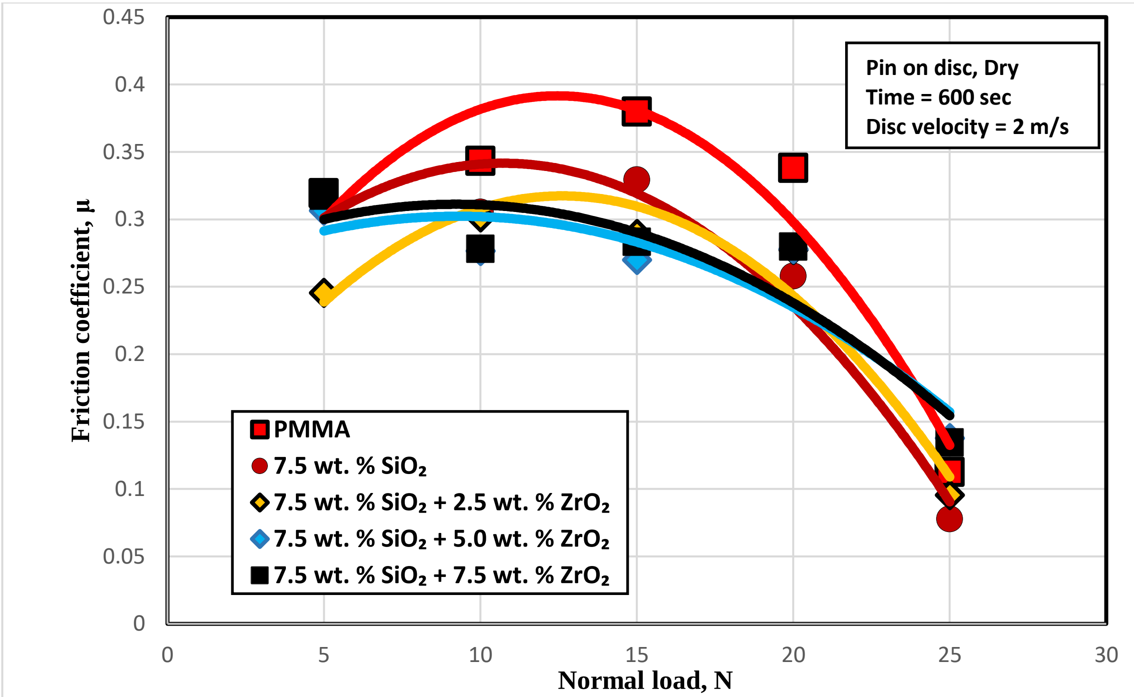
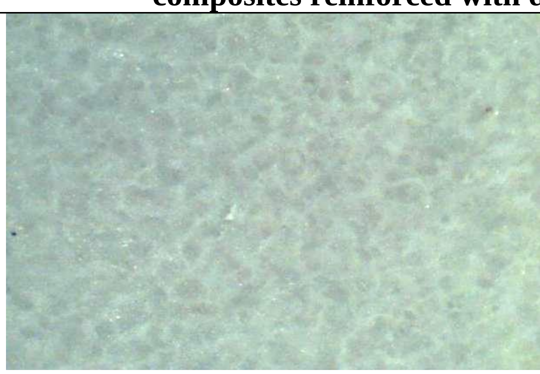
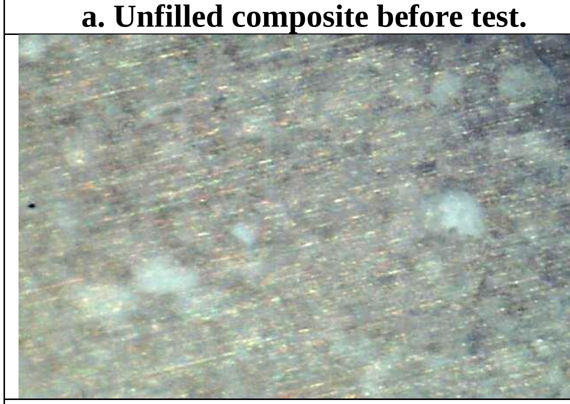


Fig. 9 The relation between normal load and average friction coefficient for heat cured unfilled PMMA and (PMMA/7.5 wt. % SiO_2 nanoparticles) composites reinforced with different ZrO_2 nanoparticles content.



a. Unfilled composite before test.

b. Unfilled composite after test.



c. 7.5 wt. % silica composite before test.

d. 7.5 wt. % silica composite after test.

Fig. 10 Photomicrographs of unfilled PMMA and PMMA/7.5 wt. % SiO_2 nanoparticles composites before and after test.

Wear of heat cured unfilled PMMA, PMMA reinforced with7.5 wt. % of silica nanoparticles and PMMA reinforced with (7.5 wt. % SiO_2 and (2.5, 5 and 7.5 wt. % ZrO_2)) nanoparticles is shown in Fig. 8. It is clear that, with the increment of the applied normal load the wear increases due to the increase of the contact area. It is also obviously seen that, the maximum wear value was observed for unfilled PMMA and the optimum composition of the mixture was PMMA reinforced with 7.5 wt. % SiO_2 and 5 wt. % ZrO_2 nanoparticles that gives the minimum wear value. The relation between the normal load and the average friction coefficient for heat cured unfilled PMMA, (PMMA/7.5 wt. % SiO_2 nanoparticles) composites and PMMA reinforced with (7.5 wt. % SiO_2 and (2.5, 5 and 7.5 wt. % ZrO_2)) nanoparticles is illustrated in Fig. 9. The maximum friction coefficient values were observed for unfilled PMMA. By reinforcing PMMA with (7.5 wt. % SiO_2 and (2.5, 5 and 7.5 wt. % ZrO_2)) nanoparticles, the average friction coefficient decreases. The photomicrographs of unfilled PMMA and PMMA/7.5 wt. % SiO_2 nanoparticles composites before and after test are illustrated in Fig. 10. Unfilled composites suffered severe wear represented in material flow on the worn surface, Fig. 10, b. Composites filled by silica nanoparticles showed less surface damage due to the improved wear resistance of silica, Fig. 10, d.

CONCLUSIONS

1. Reinforcement of PMMA composites with SiO_2 nanoparticles improves the wear resistance for contents up to 12.5 wt. %.

2. The optimum content of (PMMA/SiO_2 nanoparticles) composites was observed at 7.5 wt. % that gives the minimum wear value.

3. For unfilled PMMA and its all contents of SiO_2 nanoparticles composites, friction coefficient increases up to maximum at 15 N, then decreases with further load increase.

4. By adding ZrO_2 nanoparticles to the (PMMA/7.5 wt. % SiO_2 nanoparticles) composites the wear resistance improved at 5 wt. % of ZrO_2 nanoparticles.

5. For all PMMA composites, the optimum weight composition was observed for PMMA reinforced with (7.5 wt. % SiO_2 and 5 wt. % ZrO_2) nanoparticles composite which gives the minimum wear value.

6. The maximum friction coefficient values were observed for unfilled PMMA. By reinforcing PMMA with (7.5 wt. % SiO_2 and 2.5, 5, 7.5 wt. % ZrO_2) nanoparticles friction coefficient decreases.

7. Relatively high friction fluctuations were observed during the test. This behavior may be due to the transfer of PMMA into steel disc and transfer back to composite surface.

8. With increasing the normal load, the wear increases for unfilled PMMA and its all composites.

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