



The Effect of Peanut and Walnut Shells powders on Tensile and Microstructure of the PMMA for prosthetic Denture

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ABSTRACT

In the current research, the heat-cured matrix material powder of PMMA was reinforced with Peanut and Walnut Shells (natural powders) which are chemically treated with 5% (w/v) (NaOH) to improve the matrix bonding (PMMA) before being used as a reinforcing powder and adding to exactly similar averages particle sizes $\leq (53\mu\text{m})$, with different weight fractions of (4, 8, and 12 wt.%). The results indicated that the Elastic modulus values reached their maximum value at (8 wt.%) when reinforced with Peanut Shells powders (1.053Gpa), while, the values of tensile strength, elongation percentage at the break, decrease as the weight fraction of Peanut and Walnut Shells powders increase and the lowest values are obtained by reinforcing with Peanut Shells powders to reach their minimum values at (12 wt.%) where the lowest values of them are (29 MPa, 2.758%) respectively. The fracture surface morphology of pure PMMA seemed to be homogenous morphology in (SEM) test, whereas the fracture surface morphology of PMMA composite reinforced by (Peanut and Walnut Shells) powders and shows a smoothness fracture surface morphology this refers to brittle to semi ductile or ductile transformation.

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1. INTRODUCTION

Biomaterials are useful in making equipment to substitute part or function of the body when needed, economically reasonable, and physiologically adequate. Diverse equipment and materials are being used in the treatment of diseases and injuries [1]. Dentures are prosthetic equipment is produced to substitute missing teeth, which is held by soft and hard tissues of the oral cavity. Dentures are made from many materials like (cellulose plastics, polyvinyl chloride [2]. In many

instances the traditional denture remains the best treatment for medical and economic reasons, a perfect denture base material must include sufficient mechanical and physical features in addition to biocompatibility there is also aesthetics, polymethyl methacrylate (PMMA) is usually employed to make denture base material because of its multiple benefits which have (low price, biocompatibility, quite simple to process, stability in the oral atmosphere, and good appearance [3]. The literature reviews provide some researches, which is done in this field, it's:

W. Abbas et al. (2010), to assess preferred properties and concentrations of *Nigella sativa* and thyme as antibiotic materials compatible with low-cost oils of denture base acrylic powder to a commercially available heat-curing acrylic resin. Pure natural oil for *Nigella sativa* and thyme with concentration (0.5%, 1%, 1.5%, and 2%) to assess transverse strength, indentation hardness, color property, residual monomer, dimensional accuracy, porosity, infrared spectroscopy, and antimicrobial sensitivity tests. The results showed a significant increase of transverse strength, the rigidity of the dental base materials, with the addition of thyme and *nigella* oil and no pores, Additives from pure natural oil recommended by *Nigella sativa* and thyme at a concentration of 1.5% to give appropriate properties and antimicrobial after acrylic resin treatment of dental base, but thyme oil showed no effect on color after treatment in relation to the *nigella* [4]. J. K. Oleiwi et al. (2013), studied the influence of the particles size and volume fraction of silica (SiO_2) ceramic particles on the mechanical properties of the PMMA polymer. The results indicate that the maximum shear stress, bending modulus, tensile strength, elongation percentage and modulus elasticity of PMMA composites are increased with increasing the addition of SiO_2 particles and with the increase in the loading filler of SiO_2 particles. The values also showed that the impact of energy and fracture toughness of the PMMA composites decreases when the volume fraction of SiO_2 particles is increased. The mechanical properties improved when reinforced with a small particle size of Silica more than a large particle size [5].

H. R. Fahimeh et al. (2014), the aim of this study is to make an in-vitro comparison of the thermal conductivity, compression strength, and tensile strength of the acrylic base of complete dentures of with those made of nanosilver reinforced acrylic (PMMA mixed with 5% Wt.) nanosilver. This study showed that the mean thermal conductivity and compressive strength of PMMA strengthened with nanosilver were significantly higher than the unmodified PMMA, while the tensile strength decreased significantly after the combination of nanosilver. The results proposing the favorable effect of silver nanoparticles on enhancing the thermal conductivity and compressive strength of PMMA [6].

A. Vipul et al. (2015), to assess and compare transverse strength, impact strength, surface hardness, and water absorption of (10% and 20%) by weight zirconia (ZrO_2) reinforced high-impact acrylic resins samples. The result was a marked increase in the transverse force in the reinforced samples when compared to the control group. Impact strength and surface hardness were found to have lower values compared to the control group. Water absorption has been found to increase the addition of (10% and 20%) zirconia (ZrO_2) [7].

S.I. Salih et al. (2015), Studied the properties of PMMA by applying two forms of particles nanohydroxyapatite and micro-zirconia with various loading filler (1, 2, and 3 vol.%). The results indicate that with increasing of the volume fractions of (nHA) and (ZrO_2) particles the tensile strength of PMMA composites (PMMA-nHA), (PMMA- ZrO_2), and hybrid laminated composites specimens will be increased [8].

J. K. Oleiwi et al. (2017), studied the PMMA properties when reinforced with a Siwak fiber and bamboo fibers with different weight fractions (3, 6, and 9 wt.%) to enhance the tensile properties. The fibers were cut to three lengths and different concentrations are used, the best results of tensile properties were obtained with the maximum fibers length and higher weight fractions [9,10].

M. A. Khan et al. (2017), studied adding zinc oxide (ZnO) to the (PMMA) to the prepared composite material and utilizing melt mixing process. PMMA composites produced with various ZnO loadings of (0, 0.5, 1, and 2 %) by weight. The results indicate that the tensile strength of the composites decreased significantly with an increase in ZnO content [11].

Z. S. Abdullah et al. (2018), enhanced acrylic resins using different fibers or fillers by weighting (0.5%, 1%, and 1.5%) of both polyester and polyamide fiber were added to acrylic cure. This study was conducted to evaluate the effect of treated fibers on the mechanical properties of basic dental materials poly (methyl methacrylate). The results show the addition of salinized treated fiber

(polyester, polyamide, and combination of both fiber) improve transverse, impact strength, and hardness properties of denture base material and has no effect on the surface roughness [12].

J. K. Oleiwi et al. (2018), investigated the effect of adding two various forms of strengthening powders, including (Rice Husk and Bamboo powders) that were practical size (25 and 75 μm) and used at four different concentrations (2,4,6, and 8 wt.%), to locate improved in the properties of tensile for heat-cure acrylic resin. The results indicated that with a low size of particles and high concentration the tensile strength and modulus of elasticity increased. Also, the greatest values of tensile strength and modulus of elasticity were obtained when reinforced with Rice Husk powder at (25 μm) and (8 wt. %) [13].

S. I. Salih et al. (2018), polymethyl methacrylate (PMMA) resin as a matrix material have been reinforced by two different natural powder in nanometre size (pomegranate peels and seed powder of dates Ajwa to prepare two groups of PMMA nanocomposites samples in individually form, with different weight fractions ratio (0, 0.4, 0.8, 1.2 & 1.6 wt. %). The result showed that the highest properties of biocomposite specimens were obtained by reinforced with pomegranate peels powder in comparison with biocomposite specimens reinforced by dates Ajwa nano seed powder [14].

2. MATERIALS AND METHODS

The composite prosthetic complete dentures specimens in this research, consisting of a matrix polymer and strengthening materials powders. The Matrix material involved heat-cured PMMA which was used as a fluid resin matrix, type (Spofa Dental Company) for preparation composite prosthetic denture base specimens. Two forms of selected natural powders with various weight fractions of (4%, 8%, and 12% wt.) and one particle size ($\leq 53\mu\text{m}$) were applied to the acrylic powder involving (Peanut and Walnut Shells) powders. Figure 1 displays the two types of natural particles after the grinding process. The base materials of PMMA denture including from powder as a polymer and liquid monomer (methyl methacrylate, MMA). The usual mixing ratio proportion for heat cure acrylic resin is typically volumetric ratio between (2.5) polymer powder (PMMA) and (1) monomer liquid (MMA). due to the solution of the polymer in the monomer, many changes can occur When combining powder and liquid. This ratio had an influence on the mixture's workability, dimensional changes, and toxicity of acrylic resin specimens [15]. by using a metallic plate with a size equivalent to the size of the mold cavity, the mold-pressed to obtain a smooth surface and to prevent gases vapor from entering PMMA during the curing was sealed in a closed container with pressure conditions of (70°C, 2.5 bar, and 30 min.) according to company instructions. Then the temperature rises to (100 °C) and stays at this temperature for an hour, to eliminate the residual monomer, the mold cooling process stars inside the curing device.

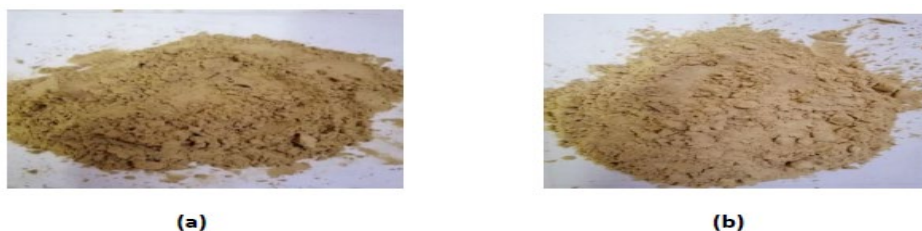


Figure 1: (a) Peanut Shells powder after grinding. (b) Walnut Shell powder after grinding.

3. EXPERIMENTAL TESTS

I. Tensile Test

The test was conducted at room temperature by using universal tensile machine type (LARYEE) in the Materials Engineering Department-University of Technology), with a load capacity of (50kN) crosshead speed was(2mm\min), and the tensile load was applied slowly until the fracture was reached. The tensile specimens are prepared according to the ASTM D638 standard. The dimensions of the specimen were (Length 150mm, Width 20mm, Thickness 5mm, Gauge length 60mm, Narrow width 10 mm).

Figure 2 displays the standard tensile test specimen. Figure 3 displays the composite specimens before and after the tensile test [16].

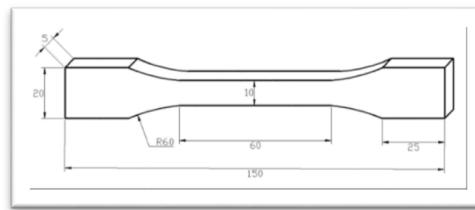


Figure 2: Tensile Test Specimen ASTM.



Figure 3: Composite Specimens Before and After Test.

4. RESULTS AND DISCUSSION

The results of the tensile strength testing performed on neat PMMA and PMMA composite specimens that prepared in this study are shown in Figure 4, this shows the effect of adding both types of natural powders which include (Peanut and Walnut) Shells as reinforcing materials at particle size about ($\leq 53 \mu\text{m}$) and different weight fractions. From Figure 4 it can be seen how the tensile strength values in both groups of PMMA composite materials decreased when the weight fraction of these powders increased. This is due to the continuity between the matrix and reinforcing powders which are in turn decreased with increasing the weight fraction of these materials and lead to material failure at lower stress then decreasing in the tensile strength of these composite materials. It can also be noticed that the presence of Peanut Shells powders has a remarkable effect on the tensile strength of PMMA composite specimens than Walnut Shells powders. Therefore, the tensile strength for composite specimens (PMMA–Peanut Shells) is lower than the tensile strength values for composite specimens (PMMA–Walnut Shells). Also, the value of tensile strength decreased from (55 MPa) for neat PMMA specimens (as referenced) to the lower value of (29 MPa) for composite materials (PMMA–12 % Peanut Shells) [17-19].

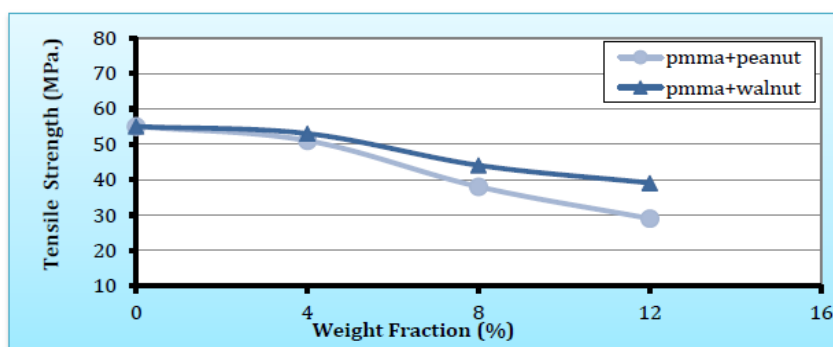


Figure 4: Tensile Strength and weight fraction of (Peanut and Walnut) Shells powders for PMMA composite specimens at the same average particle size ($53 \mu\text{m}$).

The Elastic modulus values results that performed on neat PMMA and PMMA composite specimens for prosthetic denture base materials that prepared in this study are shown in Figure 5, which show the effect of adding natural powders which include (Peanut and Walnut) Shells powders at particle size ($\leq 53 \mu\text{m}$) and different weight fractions on the elastic modulus of PMMA composite. From this Figure, it can be seen the elastic modulus increased by increasing the weight fraction of

these powders and it is reaching the maximum values at (8 % Wt.). This is because the strengthening mechanism which meaning these powders minimized the slippage of the PMMA chains by filling the spaces within the PMMA matrix and producing a complete interface, that formation of high strength bonding between these powders and the PMMA matrix. In addition, the nature of these natural powders has high elastic modulus and high stiffness when compared with PMMA resin so the total composite stiffness will be increased. Also, the elastic modulus values of PMMA composite specimens will decrease when the weight fraction of (Peanut and Walnut) Shells powder exceed (8 %wt.) but still higher than the value of the referenced specimen. This is due to the lack in the wettability and weakly the physical bonds between the matrix and these powders when increasing the weight fraction of these powders to reach the supersaturated state that tends to aggregate these powders and void formation from the entrapped air and moisture that weakening these composite materials. The addition of peanut shells particles has a remarkable effect on PMMA composite specimen's Elastic modulus more than walnut shells particles, therefore, the Elastic modulus for composite specimens (PMMA–peanut shells) is higher than the Elastic modulus values for composite specimens (PMMA– walnut shells). Thus, the Elastic modulus value for PMMA specimen (as referenced) increased from (0.848 GPa) to reach the higher value of (1.053 GPa) for composite materials (PMMA–8% peanut shells) [20].

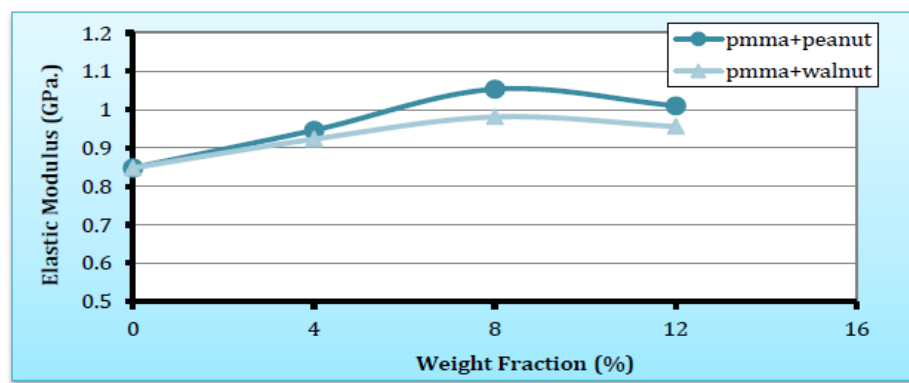


Figure 5: Elastic modulus and weight fraction of (Peanut and Walnut) Shells powders for PMMA composite specimens at the same average Particle Size ($53 \leq \mu\text{m}$).

The elongation percentage values for PMMA that prepared in the current study are shown in Figure 6, which demonstrated the relationship between the elongation percentage and the weight fractions of (Peanut and Walnut) shells powders. It can be seen that the effect of adding these powders at particle size about ($\leq 53 \mu\text{m}$) and different weight fractions on the elongation percentage of PMMA composite material, the elongation percentage values decreased with an increase of the weight fractions of these powders in PMMA composite materials. This behavior causes the increasing in the number of particles and will act as localized stress concentration regions that decreasing the elongation percentage. Also, the particles of these powders entering the resin chains which plays the role of restricting the movement of the chains. So, the slippage of these chains was decreased and this led to decreasing the elongation percentage. It can also be noticed that the addition of Peanut Shells powders has a remarkable effect on the elongation percentage of PMMA composite specimens more than Walnut Shells powders. Therefore, the elongation percentage for composite specimens (PMMA– Peanut Shells) is lower than the elongation percentage values for composite specimens (PMMA– Walnut Shells). Thus, the elongation percentage value for composite materials (PMMA–12% Peanut Shells) is decreased from (8.1 %) for neat PMMA specimen (as referenced) to reach the lower value of (2.758 %) [17,21,22].

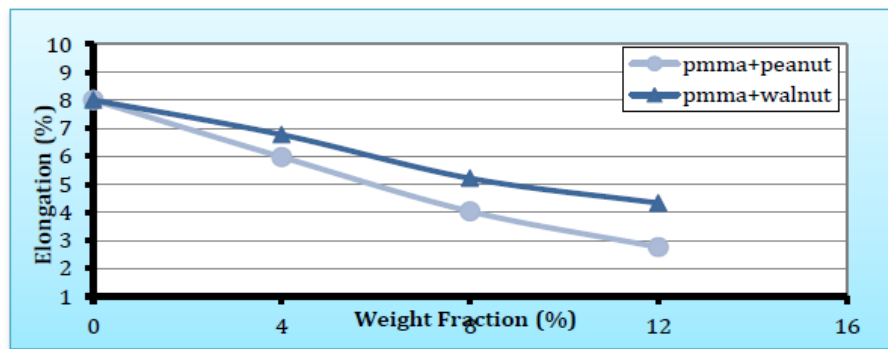


Figure 6: Elongation (%) and weight fraction of (Peanut and Walnut) Shells powders for PMMA composite specimens at same Average Particle Size ($53 \leq \mu\text{m}$).

Scanning electron microscopy (SEM) has been done to fit the mechanical behavior and the morphology of fracture surface for pure PMMA and PMMA composite specimens, that is reinforced with (Peanut and Walnut) shells powders at average particle size ($\leq 53\mu\text{m}$) and specific weight fractions of (4, 8 and 12%wt.). The morphology of polymeric composite depending extensively on a variety of factors such as the manufacturing conditions, particle size, components nature, product design, and components ratios. Figure 7: a and b show the fracture morphology of pure PMMA at different magnifications (500x and 1000x), which appears a Brittle failure of homogeneous material.

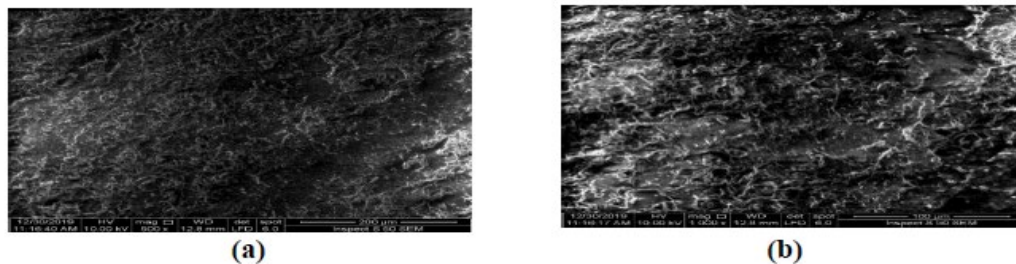


Figure 7: Represents SEM Image of Fractured Surface for Pure PMMA at Different Magnification(a) 500x and (b) 1000x.

Figures 8-10 respectively display the surface morphology of fracture the PMMA composite samples strengthened by (4, 8, and 12%) of Peanut Shells at various magnifications (500x and 1000x). This is identified by microscopic Imaging of SEM which revealed a heterogeneous morphology. Figure 8 a, b shows the presence of some small particle damage that stretches back to the destroyed and fragmented particles when the breakdown occurred in the sample. It can be also appearing that the damaged particles remain covered with material, also after failure, all these Figures indicates a semi smoother to smoother fracture surface at microstructure morphology, and semi-continuous morphology which tends to be embedded Peanut Shells powder in PMMA matrix and show the good distribution of these powders in PMMA matrix. This is appearing significant variation in the behaviors for the fracture surface morphology when reinforcing with Peanut Shells powders as compared with the behavior of pure PMMA matrix. Also, increase the smoothness that is lead to suggests transformation from brittle to ductile or semi-ductile fracture in the matrix material.

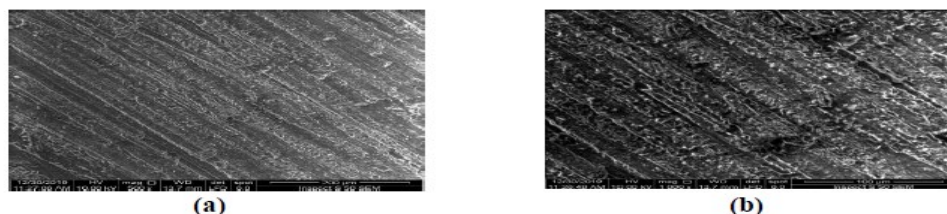


Figure 8: Represents SEM Image of PMMA Composite Reinforced with (4 % wt.) Peanut Shells Powder for Average Particle Size $\leq (53\mu\text{m})$ at Different Magnification (a) 500x and (b) 1000x.

Although, Figures 9 and 10 at higher magnifications of the fracture morphology of PMMA composites specimen equal to (1000x) the individual microparticles are seen sitting in a tiny void without any connection of polymer. This means that the dewetting of the particles from the matrix happened both before or during the mechanical test. The voids often tend to be elongated, in a specific direction, indicating a form of energy dissipation that exists in these high strains of composites failure [23].

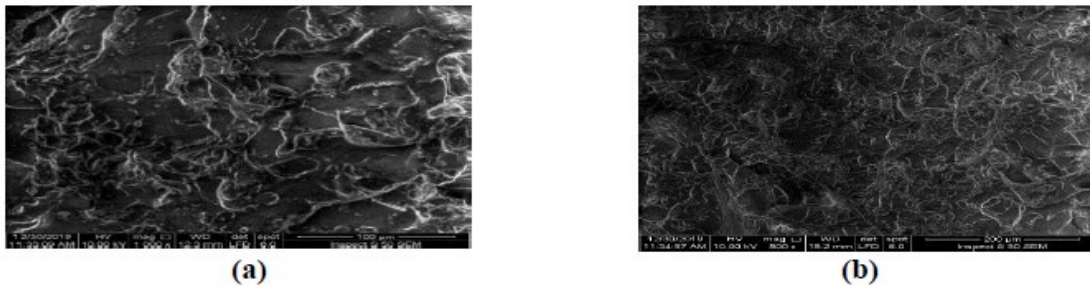


Figure 9: Represents SEM Image of PMMA Composite Reinforced with (8 % wt.) Peanut Shells Powder for average Particle Size $\leq(53\mu\text{m})$ at Different Magnification (a) 500x and (b) 1000x.

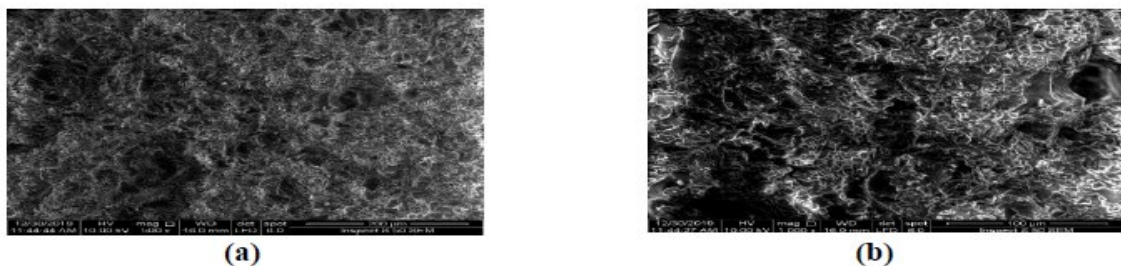


Figure 10: Represents SEM Image of PMMA Composite Reinforced with (12 % wt.) Peanut Shells powder of average Particle size $\leq(53\mu\text{m})$ at Different Magnification (a) 500x and (b) 1000x.

The morphology of fracture surface of PMMA composite specimens reinforced with Walnut shells powder at the same average particle size in PMMA matrix at various magnifications (500x and 1000x) as seen in Figures 11- 13 respectively. It has been noted that from these figures the smooth fracture surface morphology and semi-continuous morphology also indicate a reasonable distribution and embedded of these powders in the PMMA matrix. Meanwhile, Walnut Shells particles are relatively good and the particles are incorporated in the matrix and have become part of it essentially, there is a good and uniform dispersion exist throughout the entire PMMA matrix that is resulting from the strong interaction and interfacial contact between the PMMA matrix and Walnut Shells powder in the composite specimens. There is a significant variation in the behavior of the surface morphology of fracture of pure PMMA relative to the PMMA composite reinforced with Walnut Shells powder at the same particle size, where the smoothness increases and this means a brittle to ductile or brittle to semi ductile transformation. Even though Figure 13 indicates that the fracture surface tends to be fibrous form this is referred to ductile fracture with high strength material. All these figures in this test do not display any density gradients in the cross-section analyzes of fracture reason for PMMA composites specimens. thus, it is displaying a homogenous microstructure in all specimens. In addition, the results in these composite materials revealed a strong match between the PMMA matrix and reinforcing materials (Peanut and Walnut) Shells powders. Moreover, no attributes to the creation of cracks development growth inside the PMMA matrix were detected from these figures [24].

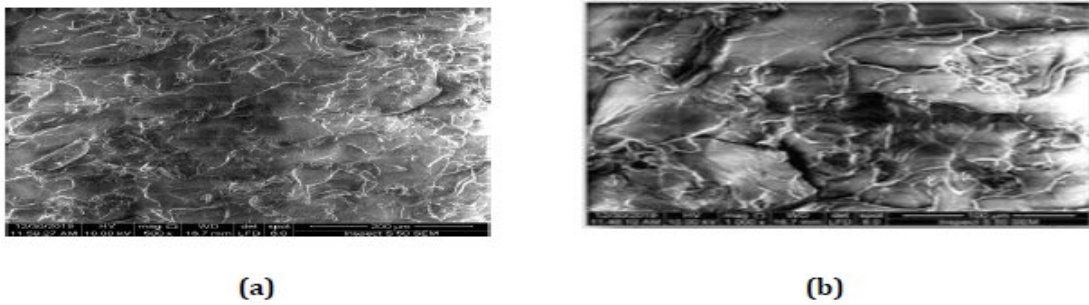


Figure 11: Represent SEM Image of PMMA Composite Reinforced with (4 % wt.) Walnut Shells powder of average Particle size \leq (53 μ m) at Different Magnification (a) 500x and (b) 1000x.

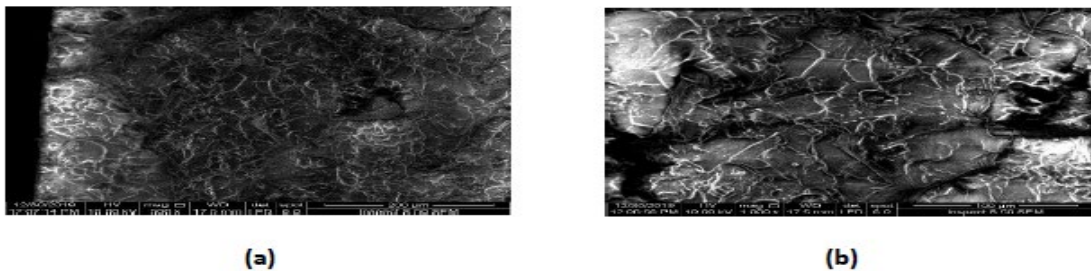


Figure 12: Represents SEM Image of PMMA Composite Reinforced with (8 % wt.) Walnut Shells powder of average Particle size \leq (53 μ m) at Different Magnification (a) 500x and (b) 1000x.

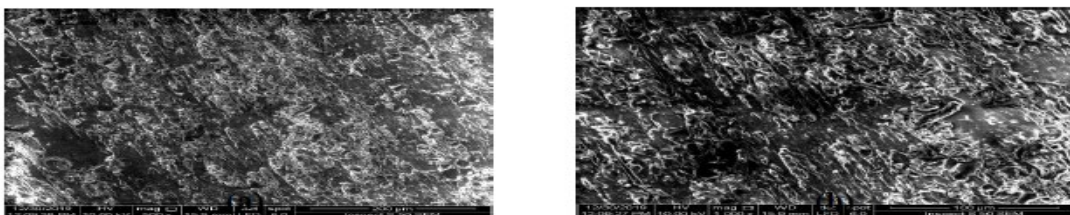


Figure 13: Represents SEM Image of PMMA Composite Reinforced with (12 % wt.) walnut Shells powder of average Particle size \leq (53 μ m) at Different Magnification (a) 500x and (b) 1000x.

5. CONCLUSIONS

- 1) The tensile strength and elongation percentage properties were decreased by increasing the weight fractions of both reinforcing powders (Peanut and Walnut) shells powders in PMMA resin. The lowest values were obtained for these properties by adding Peanut shells powder at (12 wt.%) and they were (29 MPa, 2.758 %) respectively.
- 2) The Elastic modulus was increased by increasing the weight fractions of both reinforcing powders (Peanut and Walnut) shells powders in PMMA resin. The highest value was obtained by adding Peanut Shells powder at (8 % wt.) and it was (1.053 GPa).
- 3) The fracture surface morphology of pure PMMA seemed to be homogenous morphology in the (SEM) test. Whereas the fracture surface morphology of PMMA composite reinforced by (Peanut and Walnut Shells) powders seemed to be semi-continuous and no density gradients morphology, although there was good compatibility between PMMA resin and reinforcing natural powders. And indicate the semi ductile fracture occurs.

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