



INVESTIGATION OF MECHANICAL PROPERTIES OF PMMA COMPOSITE REINFORCED WITH DIFFERENT TYPES OF NATURAL POWDERS

Sihama Issa Salih¹, Jawad Kadhim Oleiwi¹ and Arkan Saad Mohamed²

¹Material Engineering Department, University of Technology, Baghdad, Iraq

²Dijlah University College, Baghdad, Iraq

E-Mail: Sihama_Salih@yahoo.com¹

ABSTRACT

Poly Methyl Methacrylate were widely accepted material in dental and medical field due to the excellent biocompatibility and easy fabrication, however exhibit inferior mechanical properties. In current research, two groups of PMMA nano composites samples were fabricated by using hand lay-up method at laboratory temperature used for manufacturing of the maxillary complete or partial denture base. These samples consist of poly methyl methacrylate (PMMA) resin as a matrix material, was strengthen by two different natural powder in nanometresize (pomegranate peels (PPP) and seed powder of dates Ajwa (SPDA)) in individually form, with selected weight fraction ratio (0, 0.4, 0.8, 1.2 & 1.6 wt. %). Some mechanical properties and analytical physical properties (FTIR, SEM) were investigated. The result showed a considerably improvement in the values of these properties for both groups of bio composite specimens comparing with neat PMMA. All bio composite specimens reinforced with pomegranate peels powder in nanometre size showed the highest properties as compared with the bio composite specimens strengthened by nano seed powder of dates ajwa. The highest value of flexural strength, flexural modulus, max. shear stress, impact strength and fracture toughness were 114MPa, 5.124GPa, 3.562MPa, 13.75KJ/m² and 8.276 MPA.m^{1/2} respectively, for bio composite specimens reinforced with pomegranate peels powder, while for bio composite specimen reinforced with seed powder of dates Ajwa 105MPa, 4.187GPa, 3.316MPa, 10KJ/m² and 6.389 MPA.m^{1/2} respectively. On the basis of these results, it can be conclusion that the addition of natural fillings nanoparticles (pomegranate peels powder and seed powder of dates ajwa) to bio PMMA material, is one of the promising materials in use to improve the strength of fracture to the base of dental kits.

Keywords: PMMA, natural powder, nano composite, mechanical properties.

1. INTRODUCTION

A denture is a removable replacement for the surrounding tissues and missed teeth. There are two types of dentures are including: complete and partial denture bases.

Poly (methyl methacrylate) (PMMA) is the most popular material used in removable denture base material. It commonly fabricated from PMMA and it remain the most well-known of all the polymeric denture base materials due to its easy laboratory manipulation, ease in polishing and finishing, inexpensive apparatus's, stable in the oral environment, and good appearance. Therefore, Dentures made from resin based polymeric systems were more popular as compared with metal-base denture. On the other hand, one of the main problems of this material is considered to be its poor mechanical performance [1].

The most commonly problem which takes place in denture base serving is the fracture of acrylic resin, which is the basic failure of prosthodontics denture-based materials and yet remains unsolved problem [2]. In a survey, found that (63% to 68%) of complete denture prostheses had broken within few years after fabrication by impact failure outside the mouth when the denture was accidentally dropping on hard surface, and very low strength especially under fatigue failure inside the mouth by refined occlusal biting force [3]. Therefore, improvements in the mechanical performances of denture base structures have been researched by incorporating

strengthening fillers to the PMMA matrix, thus producing strengthened denture base resins.

The influence of glass fiber as a strengthening material, on fracture resistance and flexural strength of acrylic denture base resin was investigated, the result was giving an indication about the possibility for beneficent the flexural strength of heat cured PMMA after strengthening with glass fiber [4].

The influence of adding (5-20 %) micro hydroxyapatite (HA) on some mechanical properties in addition to thermal properties of heat cured PMMA resin. By the addition of HA, the flexural modulus of PMMA was increased. In addition, that the fracture toughness, young modulus and glass transition temperature of PMMA were affected by the addition of HA particles [5]. The mechanical properties of PMMA acrylic resin under influence of the volume fraction and particle size of silica (SiO₂) ceramic as a reinforcement material. The results exhibited that the bending modulus, modulus elasticity, maximum shear stress, tensile strength and elongation percentage of PMMA composites are improved with increasing the volume fraction content of SiO₂ particles. Whereas, that impact strength and fracture toughness of PMMA composites reduced with increases volume fraction content of SiO₂ particles [6]. Evaluated the influence of polyester fiber as reinforcing material on acrylic resin denture base materials. The results showed that specimens reinforced with polyester fibers enhanced the mechanical properties (compressive strength, flexural



strength, modulus of elasticity and degree of deflection) of heat cured PMMA acrylic resin and cold cured PMMA acrylic resin [7]. An improved in PMMA acrylic resin characteristics by adding four types of nanoparticles, which were zirconia, fly dust, fly ash and aluminium as a reinforcing material with different volume fractions of (1%, 2% and 3%) to self-polymerized PMMA resin. The results exhibited that the values of the flexural strength, flexural modulus, hardness and maximum shear stress improved by adding of Nano powders (zirconia, fly dust, fly ash and aluminium) [8]. Evaluated the effects of adding aluminium oxide powder on the properties of a conventional heat-polymerized acrylic resin. The result showed that the reinforcement of the conventional heat-cured acrylic resin (DPI) and high impact heat cured acrylic resin (LUCITONE) with 15% by wt.% aluminium oxide powder significantly increased its flexural strength and hardness with no adverse effects [9]. Surabhi, *et al*, compared the flexural strength property of poly methyl methacrylate resin reinforced with multiwalled carbon nanotubes (MWCNTs) and processed by conventional water bath technique and using microwave energy. The result of this study showed that the heat polymerized denture base resin with and without reinforcement of MWCNTs and polymerized by microwave technique possess higher flexural strength than heat polymerized fiber reinforced denture resin polymerized by water bath technique [10].

In present time natural fillers are most widely used in composites fabricating. The purpose of using natural fillers is to minimize the cost effective, as well as relatively high strength value to weight ratio can be obtained. From recent studies in this field that the investigated some mechanical properties of poly methyl methacrylate resin as a denture base material reinforced with natural material (siwakfibers and bamboo fibers). The result showed improvement of compression strength by increasing the weight ratios of both fibers, whereas, the compression strength reduced by increasing fiber length, at the same time the impact strength values reduced for all composite specimens [11]. Another study of the same materials showed that improvement in hardness, young modulus, tensile strength, fracture toughness with increasing of the length and content ratios of siwakfiber, whereas the elongation percentage at break and impact strength reduced with increasing of fiber concentration in composite samples [12]. Investigation the effect of addition of TiO_2 nano practical on impact strength, thermal conductivity and colour stability of acrylic resin cured by microwave in comparison to the conventional cured of heat-polymerized acrylic resin was done and the result of this study showed that the curing of high-impact acrylic by microwave had not changed the colour stability and thermal conductivity in comparison to the water bath, but

it decreased the impact strength. Also, the incorporation of 3% of TiO_2 improved the impact and the color stability, but the thermal conductivity was not changed [13].

The purpose of the current research is to determine the effect of adding different types of natural powders (pomegranate peels and seed powder of dates Ajwa) on the flexural properties, as well as, the impact strength and fracture toughness values for a self-polymerized PMMA which used for a denture base application.

2. MATERIALS AND EXPERIMENTAL WORKS

2.1 Materials used

Poly methyl methacrylate (PMMA) self-curing is the matrix material which was used in current work, as a pour type resin matrix material, manufactured by (Spofa Dental) company, multifunctional self-polymerizing acrylic. Table-1 shows the mechanical and physical properties of PMMA as obtain from spofa dental company.

Table-1. The physical and mechanical properties of neat PMMA (according to spofa dental company).

Property	Value (max or min)
Time of solubility	max. 4 min
Setting time	max. 7 min.
Brinell hardness	min. 120 Mpa.
Bending strength	min. 65.5 Mpa.
Absorbability	max. 32 mg/mm ³
Time required to prepare nontacky plastic mixture	4-6 min.
Solubility	max. 8 mg/mm ³
Resistance to impact	min. 0.40 J/cm ²

Two different kinds of nanometre sized natural powders (pomegranate peels (PPP) and seed powder of dates ajwa (SPDA)) were used in this study as strengthening fillers with weight fraction of (0.0, 0.4, 0.8, 1.2 and 1.6%) with average diameter of 53.38 nm and 93.78nm respectively, it was added to PMMA. The Atomic force microscope AFM was used to determine the average diameter of nanoparticle and its distribution. Figures (1) and (2) show the particlesize and distribution for pomegranate peel powder and seed powder of a dates Ajwa nanoparticles respectively, the particle size test method was mentioned elsewhere [14]. Figures (3 (a, b)) show pomegranate peels powder and seed powder of dates Ajwa respectively, were used in current research.



Avg. Diameter:53.38 nm			<=10% Diameter:35.00 nm			<=90% Diameter:65.00 nm		
<=50% Diameter:50.00 nm								
Diameter(nm)<	Volume(%)	Cumulation()	Diameter(nm)<	Volume(%)	Cumulation()	Diameter(nm)<	Volume(%)	Cumulation()
20.00	0.58	0.58	45.00	9.22	19.31	65.00	17.00	89.91
30.00	0.58	1.15	50.00	14.70	34.01	70.00	10.09	100.00
35.00	2.31	3.46	55.00	16.43	50.43			
40.00	6.63	10.09	60.00	22.48	72.91			

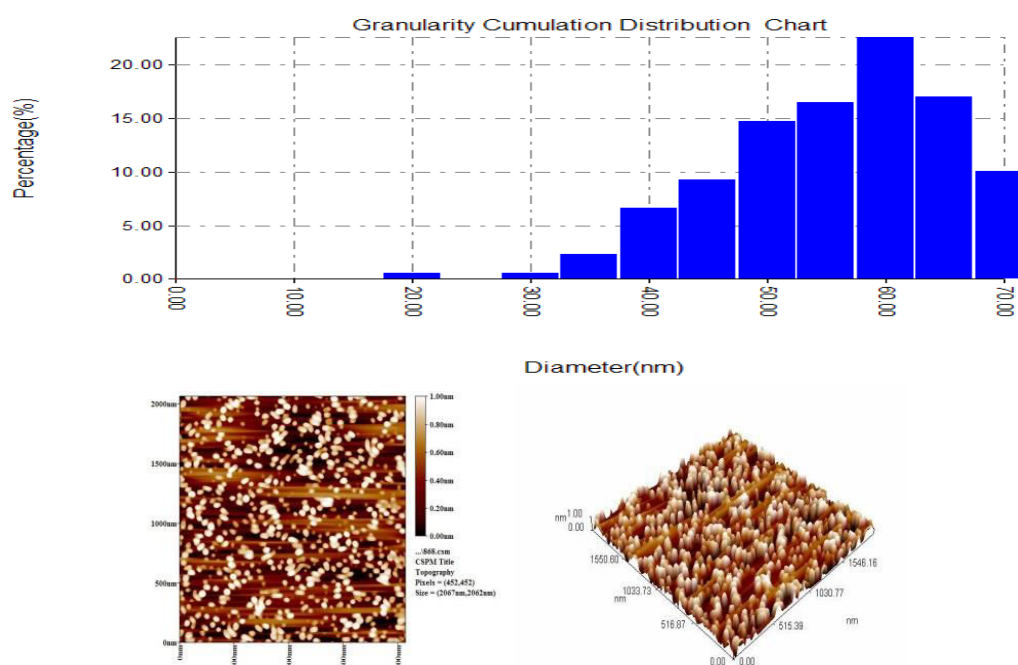


Figure-1. AFM test of pomegranate peels nanoparticles (Average diameter 53.38 nm).



Avg. Diameter:93.78 nm

≤10% Diameter:65.00 nm

≤50% Diameter:90.00 nm

≤90% Diameter:120.00 nm

Diameter(nm)<	Volume(%)	Cumulation(%)	Diameter(nm)<	Volume(%)	Cumulation(%)	Diameter(nm)<	Volume(%)	Cumulation(%)
50.00	1.17	1.17	80.00	4.09	26.32	110.00	9.94	73.68
55.00	1.75	2.92	85.00	9.36	35.67	115.00	6.43	80.12
60.00	2.92	5.85	90.00	9.36	45.03	120.00	7.02	87.13
65.00	4.09	9.94	95.00	7.60	52.63	125.00	5.85	92.98
70.00	5.26	15.20	100.00	5.26	57.89	130.00	6.43	99.42
75.00	7.02	22.22	105.00	5.85	63.74	135.00	0.58	100.00

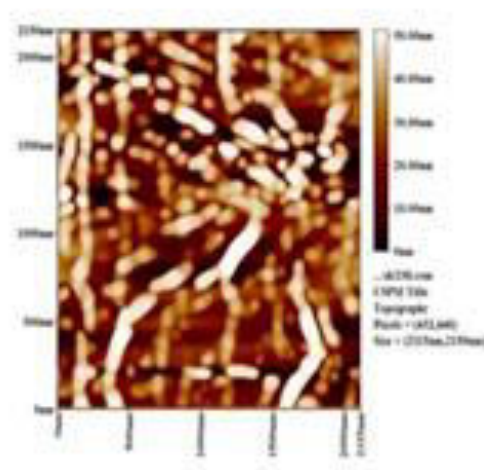
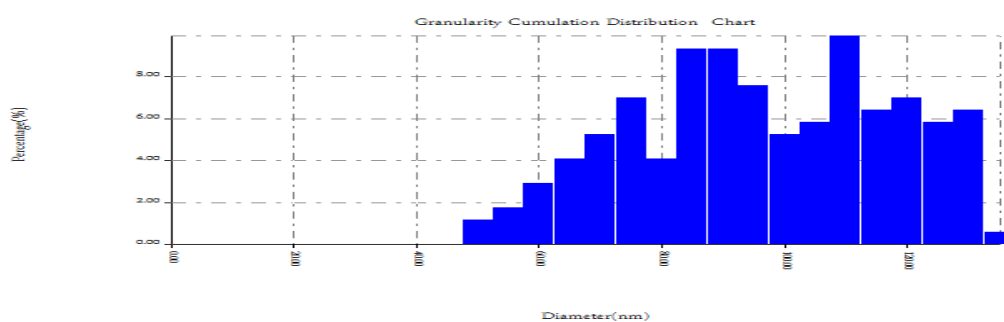


Figure-2. AFM test for seeds of a date Ajwa nanoparticles (Average diameter 93.78 nm).



Figure-3. Reinforcement materials where: (a) Pomegranate peels powder material nanoparticles size (53.38 nm) and (b): Seed powder of dates Ajwain nanoparticles size (93.78 nm).

2.2 Preparation specimens

According to the amount of PMMA acrylic resin required for filling the metallic mould cavities, the weights of the liquid monomer resin (MMA) and acrylic powder (PMMA) were estimated according to Spofa Dental Company and by using electronic sensitive balance with accuracy (0.0001) digits. The standard mixing ratio for self-cure PMMA acrylic resin, DURACRYL PLUS (self-curing base resin) is mixed in the volumetric ratio 3:1 (three parts of powder, 1 part of liquid) according to Spofa Dental Company. The importance of this ratio was related to control the workability of the mixture and dimensional changes on setting. The mixture poured with thin straight line in the centre of the metallic mould cavity. The inner and upper faces of the mould were covered by a thin layer of thermal material having smooth surface, to obtain the samples with smooth surfaces area.

The preparation of composites specimens was done according to the required selection ratio of the weight fractions of the reinforcement materials (pomegranate peels powder and powder of seeds of a dates Ajwa in nanometre size) were calculated depending on the total weight of the composite's materials ((acrylic powder (PMMA) + liquid monomer (MMA): X% of nature powders) required for filling the mould cavities by using rule of mixture.

The curing process of acrylic was performed by placing the metal mold filled and sealed inside the curing device at a temperature equal to 55 °C and pressure equal to around 2.5 bar for 30min according to company instructions. The closed mould remained inside curing

device for (30 min) in order to complete polymerization process for acrylic specimens and to improve physical properties. The characteristic of this process is the residual monomer, which must be at minimum level and the polymerization process might be completed in short time, this curing process was done inside the Multi Cure System (Ivo met) as shown in Figure-4. After a completed curing and cooling of bio composites specimens, the composites samples were removed them from the metallic mould, with very smooth of surfaces. Then, final heat treatment at 55°C for 3 hr was done to remove residual stresses as a result of de-molding of the specimens from the metallic mold cavity.



Figure-4. The locally product curing device.

3. MECHANICAL AND PHYSICAL TESTS

Fourier transform infrared spectra (FTIR) test: Fourier transformation Infrared spectrum is used to obtain specific information about the chemical bonds and molecular structure of polymer samples. The FTIR test completed according to the international measurements (ASTM E-1252) [15], by using a spectrometer of the (Fourier infrared)) manufactured by Bruker Optics company type (TENSOR-27). Infrared spectrum was used within range of (400- 4000) cm^{-1} .

Morphology test: To examine the fracture surface morphology of bio nanocomposite samples, Scanning Electron Microscope (SEM), model (Tescan VEGA-SB) was used. To achieve good clarity for pictures, all samples are first sputtered with gold from the surface along the edge.

The flexural test and impact test were performed in order to evaluation of the flexural strength and impact strength properties for all the prepared bio composites samples. After completing preparation and finishing processes for bio composite specimens, as well as, before doing any test, all the specimens would be immersed in distilled water for two day at (37±1°C) according to (ADA specification., 1999, No.12), to ensure that the denture base stay in semi oral cavity environment. Also, in order to relief the residual stresses and take out the residual monomer [16].

Flexural test: Flexural strength is one of the most important properties because under flexural load, fracture resistance and stiffness of the materials can be measured. The flexural test was performed according to



the international standard (ISO 178, 2003) at room temperature by using the universal tensile test machine manufactured by (Laryee Company in china); type is (WDW-50). The strain rate (cross head speed) was (0.5mm/min) and the load was applied gradually until the fracture of the specimen occurs [17].

Impact test: The impact properties of materials represent its capacity to absorb and dissipate energies used to measure the strength of material under impact or shock loading. Impact strength is an important property for acrylic denture base materials which have tendency to fracture if accidentally dropped on the hard surface. Impact test was performed according to (ISO-180) [18], by using Izod Impact test machine type (XJU series pendulum Izod/Charpy impact testing machine). In izod impact test the specimen was clamped at one end and held vertically cantilevered beam and it has broken at impact energy of (5.5J) of pendulum and impact velocity (3.5 m/s). In this test, the samples of the impact test were done without notches [18].

4. RESULTS AND DISCUSSIONS

4.1 Fourier transform infrared spectrometers (FTIR) test result

This test is used for fully characterization of PMMA and nano-composites specimens as a function of addition nature powders in nanometre size which added in individually form to PMMA material. The FTIR spectrum in the frequency range (400-4000 cm^{-1}) was used in this study.

The structure of PMMA is $[-\text{CH}_2-\text{C}(\text{CH}_3)(\text{COOCH}_3)-]_n$. So that, the infrared spectrum of PMMA results from the group frequencies of the (C-C) and (C-H) groups of the backbone chain, the (C-C), (C=O) and (C-O) units of the ester group and the C-H units of the methyl substituent [19]. The infrared spectrum of neat PMMA shown in Figure (5) is quite similar to that reported by [20 and 21]. Peaks set in a range of (3000-2800 cm^{-1}) correspond to the (C-H) stretching bond of methyl group (CH_3). The peak at higher wave number is the asymmetric stretch bond of C-H is obtained at 2992.85 cm^{-1} and the lower peak is the symmetric stretch bond of C-H is obtained at 2850.55 cm^{-1} . The peaks around 1400 cm^{-1} are assigned to C-H aliphatic bending bond. The absorption peaks around (2992.85 cm^{-1} and 2927.30 cm^{-1}) correspond to C-H asymmetric stretching in CH_3 and CH_2 , respectively. The vibrational band at 2850.55 cm^{-1} is due to the C-H symmetric stretching in CH_3 . The characteristic band for the neat PMMA is observed at 1722.15 cm^{-1} , which corresponds to C=O stretching band. The vibrations due to deformation modes of CH_3 groups appear at 1482.14 cm^{-1} , at 1434.55 cm^{-1} and at 1385.75 cm^{-1} . Medium bands at (1269.82 cm^{-1} and at 1239.04 cm^{-1}) correspond to C-O stretching modes. The band at (1189.91 cm^{-1}) corresponds to CH_3 wagging, and two bands at (1143.15 cm^{-1}) are due to the CH_3 twisting. The vibration modes due to C-C stretching appear at (985.66 cm^{-1} and 965.14 cm^{-1}). The peaks at (911.13 cm^{-1} and 840.69 cm^{-1}) are assigned to CH_2 rocking, and the peaks

at (810.42 cm^{-1} and 749.14 cm^{-1}) are due to the CH_2 rocking in plane and out of plane bending, respectively [20 and 21].

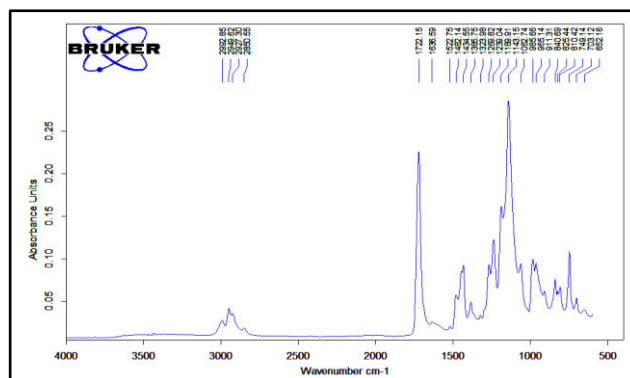


Figure-5. FTIR spectrum that obtained for the cold cure PMMA specimen.

Figure (6) shows the FTIR spectrum of pomegranate peel powder (PPP). The spectrum confirmed the complex nature of the peels and proved the presence of wide variety of compounds. It has been reported by studies that pomegranate peels contain different natural compounds with biological nature [22]. From this figure it can be observed that the spectra for PPP showed long bandwidth 3435 cm^{-1} which indicates the O-H stretching band confirms the presence of alcohols compounds and carboxylic acids. The C=C stretching band of alkyne group was detected at bandwidth 2924-2854 cm^{-1} . The sharp mid-intense peak at 1718 cm^{-1} attributed to carbonyl group C=O which lead to presence of aldehydes, ketones and carboxylic acids. The moderate sharp peak at 1618 cm^{-1} indicates the presence of unsaturated compounds (alkenes). The bands at 1446 cm^{-1} for $-\text{O}-\text{CH}_3$ deformation. The band at 1363 cm^{-1} (CH_2 bending), related to the presence of cellulose. A peak at 1223 cm^{-1} ($-\text{CH}_3\text{CO}$ stretching) confirms the presence of esters and ethers. A peak at 876 cm^{-1} for ($-\text{CCH}$ and $-\text{COH}$ bending, from Figure (6) it can observe that the spectra of PPP is quite similar to that reported by [23 and 24].

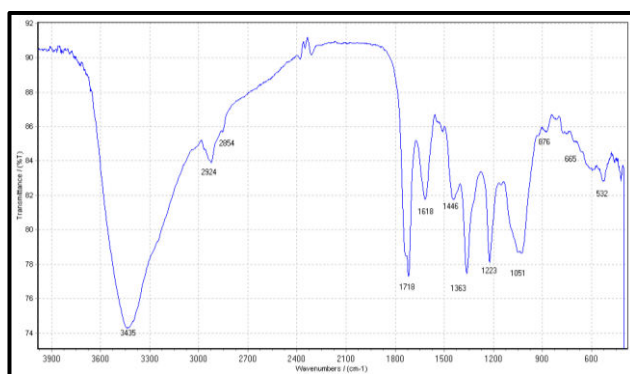


Figure-6. FTIR spectrum of pomegranate peels powder.

The infrared spectrum of PMMA which was reinforced with different ratio of nano-pomegranate peels



powder (PMMA: x% nano-pomegranate peels) as a first group of nanocomposite materials are shown in Figure (7). All the characteristics vibration bands of neat PMMA were presented in FTIR spectrum of the first group composites specimens. In addition, it can be observed that, from the infrared spectrum for the first group composite specimens (Figure-7), there is no any new peak, or peaks shifts in the spectrum of PMMA composite specimens, this is due to find physical bond and absence any cross linking in these specimens. There is a clear increase in peak intensity for all of characteristic peak with increasing Nano- pomegranate peels ratio in the composite and it reaches to maximum at 4% of nano powder of pomegranate peels, except at a ratio of 1% of the peel's powders, it was observed a decreased in peaks intensity was occur.

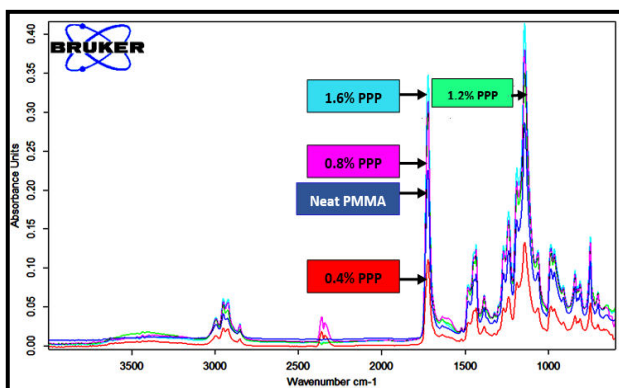


Figure-7. FTIR spectrum of (PMMA: X%PPP) polymer composite as a function of pomegranate peels powder content in composite.

Figure-8 shows the FTIR spectrum of Seed powder of dates Ajwa(SPDA). From this figure it can be observed that the FTIR spectrum of the Seeds powder of a dates Ajwa (SPDA) is very similar to the spectrum of pomegranate peels powder, since the two articles are from natural origin. The characteristics of the bands observed at 3437 cm^{-1} which indicates the O-H stretching band confirms the presence of alcohols compounds and carboxylic acids. The sharp mid-intense peak at 1716 cm^{-1} attributed to carbonyl group $\text{C}=\text{O}$. The moderate sharp peak at 1622 cm^{-1} indicates the presence of unsaturated compounds (alkenes). The infrared spectrum of SPDA (Figure-8) is quite similar to that reported for many different natural compounds by [25-27].

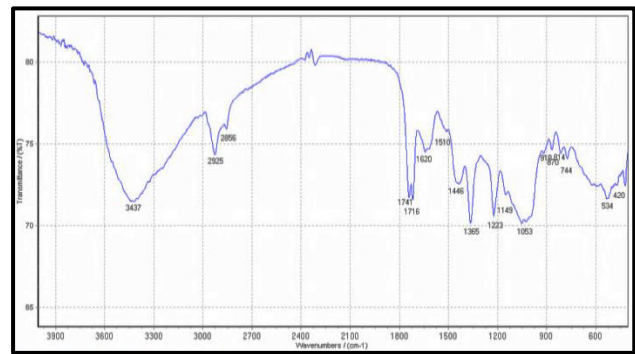


Figure-8. FTIR spectrum of (PMMA: X%SPDA) polymer composite as a function of seed powder of dates Ajwa content in composite.

The infrared spectrum of neat PMMA and second group composites specimens (PMMA: x% Seeds powder of a dates Ajwa), which was reinforced with different ratio of Nano- Seeds powder of a dates Ajwa (SPDA), are shown in Figure-9. All the characteristics vibration bands of a neat PMMA spectrum was present in (FTIR) spectrum for the second group of composites specimens. In addition, from this spectrum there are no other new peaks or peak shifts were observed for the PMMA nano composite specimens, this is due to find physical bond and absence any cross linking in these specimens. There is a clear decrease in peak intensity for all of characteristic peak with increasing seeds powder of a dates Ajwa ratio and it reaches to minimums at 0.8% Nano-powder. Then peak intensity was increased with increased Nano-powder ratio to 1.2%.

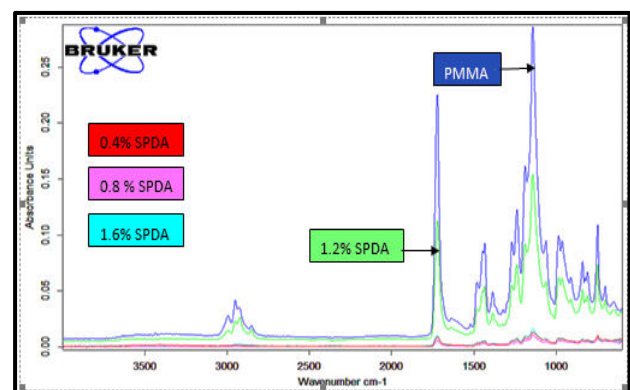


Figure-9. FTIR spectrum of (PMMA: X%SPDA) polymer composite as a function of seeds powder of a dates Ajwa content in composite.

4.2 Mechanical test result and discussion

The effect of the addition two types of natural fillers (pomegranate peels powder (PPP) or Seeds powder of a dates Ajwa (SPDA)) in individually form as a reinforcement particle to PMMA on the flexural strength, flexural modulus and maximum shear stress of PMMA nanocomposites, is shown in Figures (10, 11 and 12) respectively.

The values of flexural strength, flexural modulus and maximum shear stress for neat PMMA specimens



were equal to 78 MPa, 3.4042 GPa and 2.531 MPa respectively. When the PMMA specimen was reinforced with PPP or SPDA, it's found there is a considerably improvement in the values of this properties and for both groups of bio composite specimens comparing with neat PMMA. Afterward, a gradual increase of flexural strength, flexural modulus and maximum shear stress was observed with a further increase of the weight fraction content, for both types of a natural fillers powders that incorporated in the PMMA matrix, beside that the maximum values of flexural strength, flexural modulus and shear stress was reached at the ratio of weight fraction content (1.6%) in bio composite specimens, except the flexural modulus values of bio composite specimens which reinforced by dates seeds powder was reached to maximum values at 1.2% ratio of the fracture weight content of natural fillers. This is due to compatibility between PMMA matrix and natural reinforcing fillers [28]. Also, due to the ability of these natural particles to prevent the propagation of cracks inside PMMA matrix according to reinforcing mechanism in addition to the strong bonding between the PMMA matrix and these particles. The presence of the compatible between natural reinforcing particles and PMMA resin had a very considerable effect on a decrease in the molecular motion and reduces free volume in the prepared composites samples, which then increasing from the flexural strength, flexural modulus and shear stress properties for the prepared bio composite specimens [29].

Also, it can be noticed from these figures that the value of flexural strength, flexural modulus and maximum shear stress for bio composite specimens reinforced with pomegranate peels powder are greater than those reinforced with seeds powder of ajwa dates. This is due to the good compatibility between PMMA matrix and pomegranate peels particles and the nature of chemical composition and mechanical properties for pomegranate peels powder [30].

The highest value of flexural strength, flexural modulus and max. Shear stress were (114MPa), (5.124GPa) and (3.562MPa) respectively, for bio composite specimens reinforced with PPP, whereas for bio composite specimen reinforced with SPDA reached to (105MPa), (4.187GPa) and (3.316MPa) respectively.

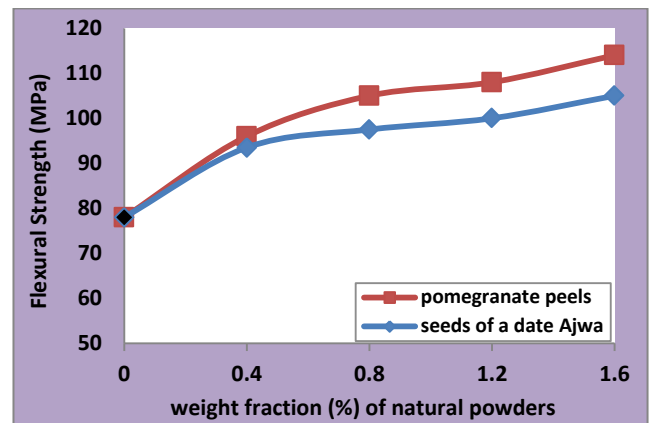


Figure-10. Flexural strength for PMMA bio composite specimens as a function of weight fraction content for PPP or SPDA in composites.

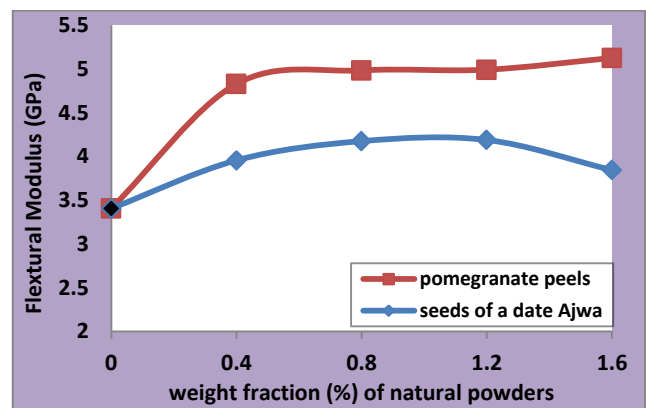


Figure-11. Flexural modulus for PMMA bio composite specimens as a function of weight fraction content for PPP or SPDA in composites.

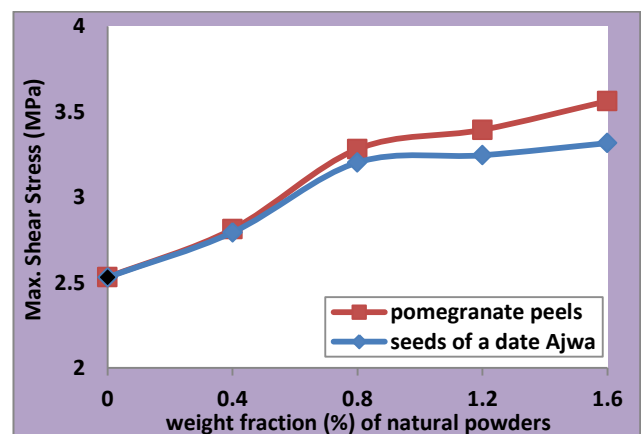


Figure-12. Max. shear stress for PMMA bio composite specimens as a function of weight fraction content for PPP or SPDA in composites.

The impact strength value is the most important property because it is giving an indication about the measure of a given material's toughness. The values of impact strength and fracture toughness for the neat PMMA and PMMA bio composite materials are showed in Figures



(13) and (14) respectively. The impact strength of composites is controlled mainly by two factors: first, the capability of the natural fillers to absorb energy that can stop crack propagation and second, poor interfacial bonding which produce micro-spaces between the natural fillers and the matrix, resulting in easy crack propagation [31].

It can be observed from these figures that the addition of the pomegranate peels and dates seeds individually, as particle reinforcing cause improvement in the impact strength and fracture toughness values of the PMMA bio composites. The reason behind this behaviour may be depend on that the impact test is a measure of a given material's toughness. So, the obtained results may be concerned with typical distribution of natural particles within the PMMA matrix and interfacial bonding between them leads to considerable increase in the energy absorbing capacity of the bio composite specimens [30, 32].

Moreover, this behaviour can be related to the obstacle and restricted the cracks from propagation inside the bio composite specimens through the addition of PPP or SPDA as nanoparticles powders to the PMMA matrix [33].

Also, it can be seen from these figures that the values of impact strength and fracture toughness for bio composite specimens reinforced with natural particles of PPP higher than bio composite specimens reinforced with SPDA. the impact strength values reach to the maximum value equal (13.75 KJ/m²) at weight fraction content of (1% and 2%) as compared with impact strength value of neat PMMA (reference specimen) which equal (8.75 KJ/m²). whereas the maximum value of impact strength for bio composite specimens reinforced with SPDA nanoparticles reached to (10 KJ/m²) at weight fraction content of (1.6%). This behaviour may be related to natural powders and good mechanical properties for pomegranate peels powder as compared with other natural fillers [34, 35].

As well as, it can be observed when comparing the fracture toughness value (Figure-14) for neat PMMA that equal 5.457 MPa.m^{1/2}, whereas, the maximum value of fracture toughness for bio composite specimens strengthened with pomegranate peels particles have particle size of 53.38 nm reaches to 8.276 MPa.m^{1/2} at weight fraction content of 0.8% and the maximum value of fracture toughness for bio composite specimens (Figure-14) strengthened with SPDA at weight fraction content of (1.2%) and for particle size (93.78 nm)reaches to 6.423MPa.m^{1/2}.

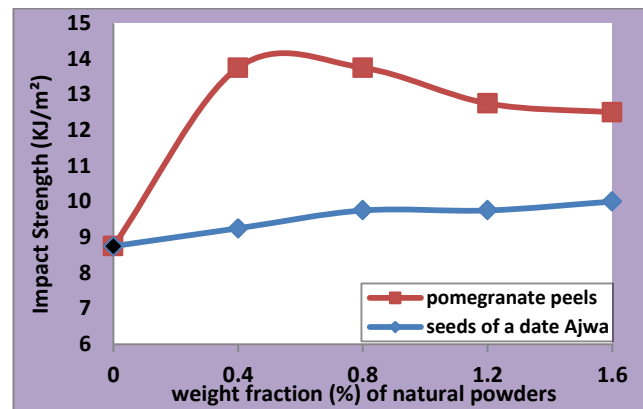


Figure-13. Impact strength for PMMA bio composite specimens as a function of weight fraction content for PPP or SPDA in composites.

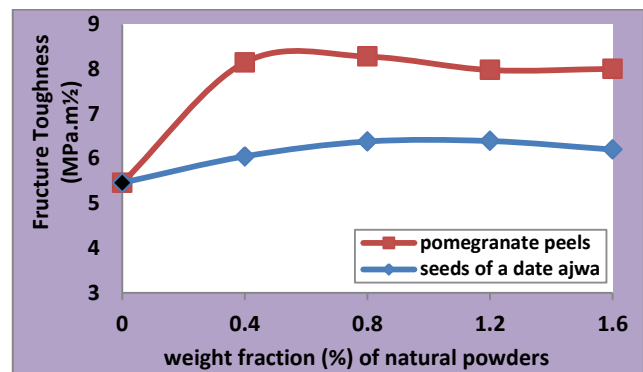


Figure-14. Fracture toughness for PMMA bio composite specimens as a function of weight fraction content for PPP or SPDA in composites.

4.3 Morphological analysis

The SEM micrographs of fracture surfaces for polymeric composites depend on many factors such as the class of polymeric material and their components and the concentration of the reinforcing materials, the wettability between their components, as well as depend on the conditions of fabrication and melting viscosity of the components [36]. In order to link the mechanical properties of PMMA bio composite with their microstructure, the scanning electron microscopy (SEM) was used. The photographic imaging for fracture surface morphology of neat PMMA (Figure-15(a)) and fracture surface morphology of composites samples which included two groups. First group samples are (PMMA: x% nano-PPP) with PPP content in composite are 0.4 and 1.6 respectively figures 15 (b and c) and second group samples are (PMMA: x% nano-SPDA) with SPDA content in composite are 0.4 and 1.6 respectively Figure-15 (d and e), all the micro graphs of sample at the same magnification (x5000). It was observed from these photographic imaging, that a homogeneous structural morphology with few micro porosities of free volume in the neat PMMA structure (Figure-15(a)). Whereas, for specimens of PMMA nanocomposites, a dense and a homogeneous structural morphology which show a



smoother fractured surface, and this indicate to the better interfacial adhesion between PMMA resin and natural nanofiller, as well as, there is no any new phase or a phase separated dominants in both groups of PMMA composites structures (Figures-15 (b, c, d and e) respectively this structural morphology appears similar with other workers [37]. Moreover, the structures of the composite substances clarify that increasing the percentage of nanoparticles content in the composite, would increase the micro structures size of the prepared composites materials. Also, the morphology of the fracture surface showed good distribution of nanoparticles, for low and high concentrations of nanoparticle content in composites, and can be observed, this through red arrows which were shown in Figures 15 (b and c) for PPP nanoparticle and 15 (d, e) for SPDA nanoparticle. As well as, through this morphology, it was noticed that most of nanoparticles (pomegranate peel powder and seed powder of dates Ajwa) are embedded inside the matrix material (PMMA), which act as an integral part of PMMA structure. Indicating to better interfacial adhesion between PMMA material and natural nano filler (PPP and SPDA). And this indicated to a good compatibility between the PMMA resin and the reinforcement nanoparticles, which enhances the mechanical properties [38 and 39].

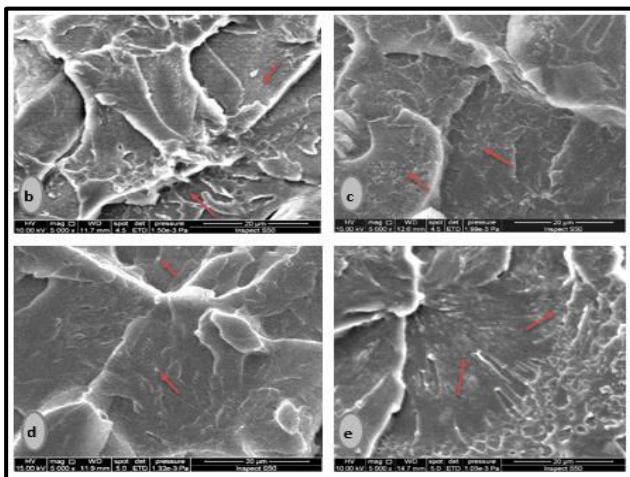
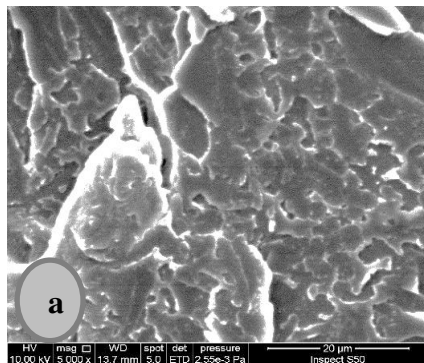


Figure-15.photographic imaging (SEM) for fracture surface morphology of: (a): neat PMMA, (b and c): Bio composites (PMMA: x% (nano-PPP) and (d and e): Bio composites (PMMA: x% (nano-SPDA). at magnification (x5000).

5. CONCLUSIONS

- Neat PMMA had the lowest value of flexural strength, flexural modulus, max. shear stress, impact strength and fracture toughness which equal (78MPa, 3.4042GPa, 2.531MPa, 8.75KJ/m² and 5.457MPam^{1/2}) respectively.
- Mechanical properties improved with adding natural powders (pomegranate peels and seed powder of dates ajwa) to neat PMMA.
- Flexural strength, flexural modulus and max. shear stress, impact strength and fracture toughness increased with increasing weight fraction content for both of natural powders (pomegranate peels and seed powder of dates ajwa) and impact strength and fracture toughness reached the highest amount for bio composite specimens when strengthened with pomegranate peels powder, at 0.8% weight fraction whereas the highest values of impact strength and fracture toughness for bio composite specimens strengthened by seed powder of dates Ajwa were at 1.6% and 1.2% weight fraction respectively.
- The highest value of flexural strength, flexural modulus, max. shear stress, impact strength and fracture toughness for bio composite specimens reinforced with pomegranate peels powder are (114MPa, 5.124GPa, 3.562MPa, 13.75KJ/m² and 8.276MPam^{1/2}) respectively. While for bio composite specimens reinforced with seeds powder of a date Ajwa, the highest values are (105MPa, 4.187GPa, 3.316MPa, 10KJ/m² and 6.389MPam^{1/2}).
- All bio composite specimens strengthened with pomegranate peels powder exhibited the highest flexural strength, flexural modulus, max. shear stress, impact strength and fracture toughness than those that reinforced by seed powder of dates ajwa at the same concentrations.
- On the basis on the mentioned earlier, it can be conclusion that the addition of natural fillings nanoparticles (pomegranate peels powder and seed powder of dates ajwa) to bio PMMA material, is one of the promising materials in use to improve the strength of fracture to the base of dental kits.

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