



INVESTIGATION THE PROPERTIES OF SILICONE RUBBER BLEND REINFORCED BY NATURAL NANOPARTICLES AND UHMWPE FIBER

Sihama I. Salih, Jawad K. Oleiwi and Hajir Mohammed Ali

Department of Materials Engineering,
University of Technology, Baghdad / Iraq

ABSTRACT

Many faces are exposed to degradation, discoloration, changes in humidity. The primary objective has improved some properties of hybrid nanocomposites materials that used for restoring of the function maxillofacial prosthesis and improving the esthetic. In the present research different lengths chopped and continuous of ultra-high molecular weight polyethylene (UHMWPE) fiber was added at selected percentage (0.0, 0.2% and 1%) to polymer blend composite (95%SR /5%PMMA: 0.2% Pomegranate Peels Powder (PPP)) for developing the properties of silicone rubber used for the maxillofacial prosthesis applications. Some mechanical and physical properties were done on the all prepared samples. The results showed that all properties have improved when added 0.5% ratio of continuous UHMWPE fiber to polymer blend nanocomposite (95%SR /5%PMMA: 0.2% Pomegranate Peels Powder (PPP)) sample and this represents the optimal ratios of components of hybrid composite sample. Therefore, this blend may be a candidate for achieving the properties required for the applications of maxillofacial prosthetics.

Keywords: Hybrid composites, UHMWPE fiber, Pomegranate peel powder, Silicone rubber.

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1. INTRODUCTION

Natural materials are able to impart certain benefits to the composite due to several advantages of it over conventional one, such as low density, low cost, ease of separation, non-corrosive, biodegradability, renewability, high stiffness, non-toxic, and environmental friendly [1]. The aim of preparing a blend of polymers is not to improve or change the main properties of the compounds but to utilize on the blend performance, processability, and uniformity of product [2,3]. Maxillofacial prostheses represent serve as replacements for a missing tissue or organ in an organism. [4]. Maxillofacial related with facial structures by artificial substitutes that movable and includes prosthetic rehabilitation of oral or facial trouble which may be obtained naturally or from disease or trauma [5]. The aim of maxillofacial prostheses is to solve the lost aesthetic look and should solve social damages problems [6].

In the last decade, there are more uses of biomaterials in order to improve the facial anatomy are silicones, polymethylmethacrylate, polyvinyl chloride, polyethylene, and polyurethane [7]. During the last five decades, silicone elastomers have been clinically choice for fabricating a facial prosthesis compared to other materials in offering optimal overall properties for facial prosthesis material. Despite the advances in material technology, the disadvantages of materials still exist such as discoloration at the edges of the prosthesis after one year of used and rupture of the silicone material due to repeated placement [8, 9]. So that many attempts were introduced to overcome these problems by development of a new class of elastomer material such polymer blends manufacturing which match some very important properties of polymers along with diminishing their individual disadvantages. For high strength structure and strong bonding between the interface of filler and rubber material, small particles incorporated into the polymer to obtain a nanocomposite material, which may improve the mechanical properties and the performances of polymeric materials [10, 11].

From previous studies in this area, it was estimated the properties of RTV silicone elastomer (MDX4-4210) when reinforced with polyurethane sheets or polypropylene fibers for auricular prostheses. The result showed that the improved the mechanical properties (tear strength, tensile strength, and modulus of elasticity), also obtaining better safety and strength of prostheses [12]. Another study shows that the hardness for two types of silicone elastomers low (Premium) and high (Silasto 30) temperature vulcanizing materials when exposed to natural aging by storage in the dark for one year. The results explain that after one year of natural aging in the dark of Premium silicone hardness was increased while the hardness value of (Silasto 30) is stable through natural aging. Each types of silicone elastomers offer clinical suitable hardness values after natural aging for maxillofacial prosthesis [13]. Ahmed Nadher Jaber et al., Studied some mechanical properties of maxillofacial silicone elastomeric materials after added polyester fiber. The results showed that mean value of tear, tensile strength, surface roughness and shore A hardness for 0.25% by weight polyester fiber (2 mm length) reinforcement group increased significantly on the contrast to the other values of reinforcement groups which were gradually decreased [14]. The mechanical and physical properties of (Silicone rubber/ polymethylmethacrylate) blends with the effect of different loading level of PMMA was studied. The results showed that the blend of (90%SR/10%PMMA) represent optimum results to mechanical properties [15, 16]. The current work was an extension and development of the results of the last papers mentioned above.

In medicine, the rind of the fruit and the bark of the pomegranate tree have been used as a source of a traditional remedy against diarrhea, dysentery, intestinal parasite and prevention from the development of cardiovascular disorders, hypoglycemic, apoptotic, antibacterial, anti-inflammatory and anti-parasitic [17]. As well as, the ultra-high molecular weight

polyethylene fiber is mostly used in dental applications. Because it has good properties as compared with metal, it has high flexibility, thin and strong. Porous UHMWPE provides a basis for ocular implants and used in maxillofacial prosthetics (chin, nose, ear, etc.) [18,19]. In the present work, hybrid nanocomposites were prepared from polymer blend (silicone rubber/PMMA) reinforced with natural pomegranate peels powder (PPP) nanoparticle and ultra-high molecular weight polyethylene (UHMWPE) fiber, for maxillofacial applications was studied.

2. EXPERIMENTAL PART

2.1. Materials Used

There are two materials used to prepare binary polymer blend as a matrix for composite include RTV Silicone rubber (VST-50F) is supply from Factor II Inc., Lakeside, USA that it is consisting of two parts, one is a liquid, and another is the catalyst. Polymethyl methacrylate (PMMA) is a second material of the blend which cold curing resin Castavaria type, provided from Spofa Dental Company. The reinforcement materials for composite are Pomegranate Peels Powder (PPP) taken from pomegranate fruit (*Punica granatum*) which were supply from Saudi Arabia with particles size 102.45 nm. Other reinforcementis (UHMWPE) fibers were supplied from China such as woven fibers with particles size 114.94 nm. Theatomic force microscope AFM was used to determine the average diameter of nanoparticle and its distribution. Figures (1) and (2) show the particle size and distribution for pomegranate peel powder and (UHMWPE) fibers respectively.

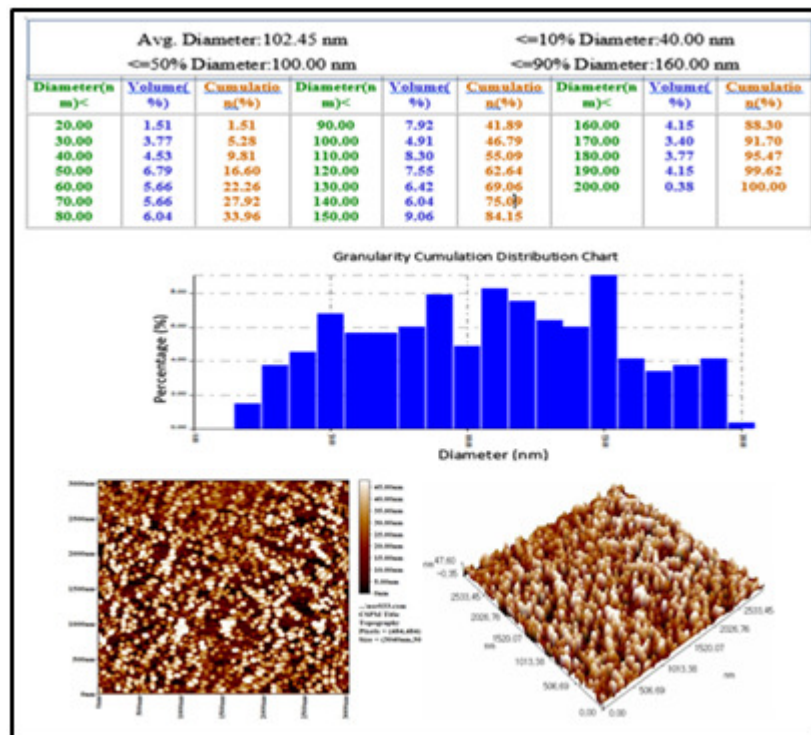


Figure (1): AFM Test of Pomegranate Peels Particles (Average Diameter (102.45 nm)).

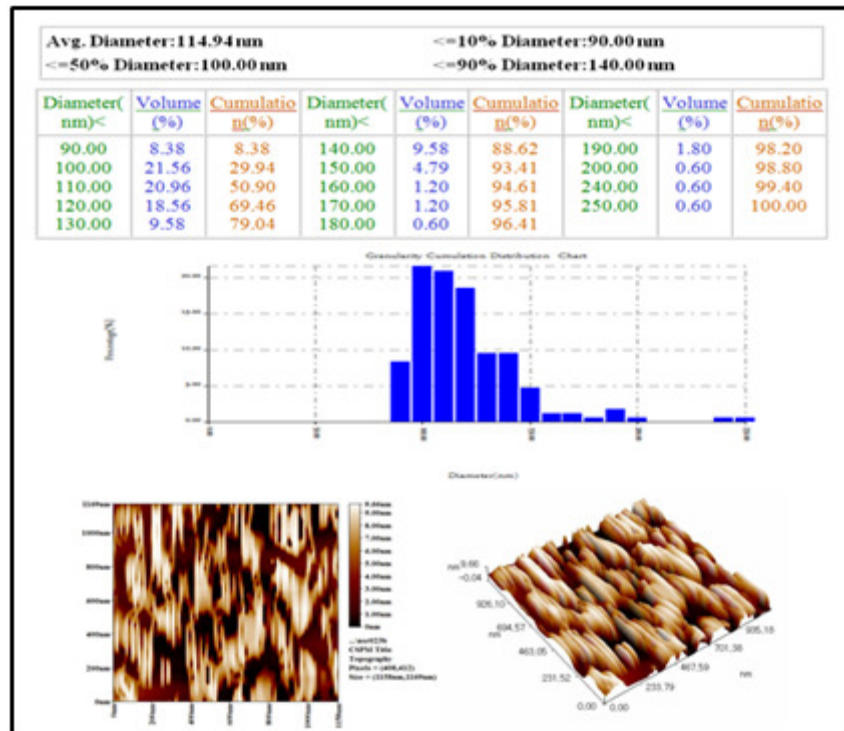


Figure (2) AFM Test of UHMWPE fibers (Average Diameter (114.94 nm)).

2.2. Preparation Method

In this work, mechanical mixing was used to prepare polymer blends composite by reinforced binary polymer blends (SR: 5%PMMA) with constant percentages ratios (0.2%) of nanoparticle (Pomegranate peels powder (PPP) and different percentages ratios (0.0, 0.5% and 1%) of UHMWPE fiber. The binary polymer blend (SR (VST-50F): 5% PMMA) prepared according to the company's instructions manufactured, the materials were mixed in the vacuum mixer for 5 minutes at speed of 360 rpm and under a vacuum of (-10 bar). Then, the blend resin pouring into the mold. Sample was left in the mold for 6 hours to vulcanization. For hybrid composites samples, the polymer blend (SR (VST-50F): 5% PMMA) were reinforced with constant percentages ratios (0.2%) of nanoparticle (pomegranate peels powder (PPP) and different percentages ratios (0.0, 0.5% and 1%) of UHMWPE fiber. Preparation of hybrid composites samples was carried out in the same practical method as in preparing the polymer blend sample mentioned above.

3. CHARACTERIZATION AND TEST METHODS

The standard ASTM E-1252 is used to perform the Fourier transform infrared spectrometer test by using the device type TENSOR-(27) made by Bruker Company (Germany) [20]. It is equipped with KBr splitter and mid-IR source (4000-400 cm⁻¹).

The tensile test of the specimens was performed according to ADA Specification No.12 and ASTM 638 standards [21, 22]. This test is done by using a universal tensile instrument with a cross-head speed of 500 mm/min.

The device Shore A hardness is used to evaluate the hardness of specimens according to the ASTM D-2240 [23, 24]. The dimension of a circular disc specimen was (6 mm) for the thickness and (25 mm) for a diameter.

The compression test of blend specimens was carried out according to the standard ASTM D-395. The dimensions of the specimen were (29 mm) for diameter and (12.5 mm) for the thickness [25, 26].

Also, the device of surface roughness is used to perform the surface roughness of specimens. The dimensions of the specimen (25x25x3) mm. Taking into consideration the average of three readings for each test [27].

4. RESULTS AND DISCUSSIONS

4.1. FTIR Results

The FTIR spectrum of neat silicone rubber (VST-50F) is shown in figure (3). The stretching vibration peak of CH_3 is assigned at absorption at 2962.78 cm^{-1} , the rocking vibration peak of $-\text{CH}_2-$ is assigned at 1413.15 cm^{-1} . While the bending peak and rocking peak of the Si-CH_3 are assigned at absorption peak at is assigned at 1258.50 cm^{-1} and 863.93 cm^{-1} respectively. The stretching vibration peak of Si-O-Si on a backbone of the neat SR assigned at 1009.28 cm^{-1} and the coupling stretching of Si-C and rocking peak of $-\text{CH}_3$ assigned at 787.08 cm^{-1} [28, 29].

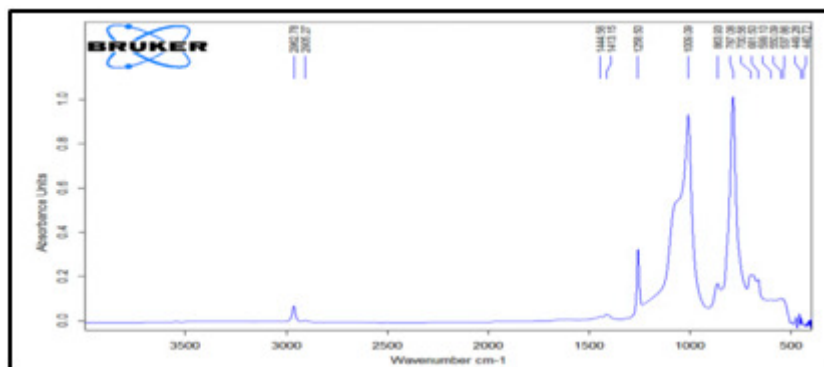


Figure 3 Represents the FTIR spectrum to the silicone rubber (VST-50F).

The FTIR spectra of pomegranate peels powder and UHMWPE were showing in figure (4, a and b) respectively. Figure (4a) illustrated that the spectrum of PPP is quite similar to that reported by [30, 31], long bandwidth 3402 cm^{-1} which indicates the O-H stretching band confirms the presence of alcohols compounds and carboxylic acids. The $\text{C}=\text{C}$ stretching band of alkyne group was detected at bandwidth 2929 cm^{-1} . The sharp mid-intense peak at 1726 cm^{-1} attributed to carbonyl group $\text{C}=\text{O}$ which lead to presence of aldehydes, ketones and carboxylic acids. The spectrum of UHMWPE fiber (figure 4b) indicates to the two sharp high intensity peaks important bands assigned at 2914.15 and 2847.48 cm^{-1} are characterized by the $-\text{CH}_2$ asymmetric stretching and symmetric stretching respectively and sharp high intensity peak at 1462.16 cm^{-1} referred to C-H bending deformation. The final two peaks at 809.68 and 716.46 cm^{-1} referred to $=\text{CH}_2$ out-of-plane bending and CH rocking mode respectively all the characteristic peaks for this spectrum are similar to that reported by [32].

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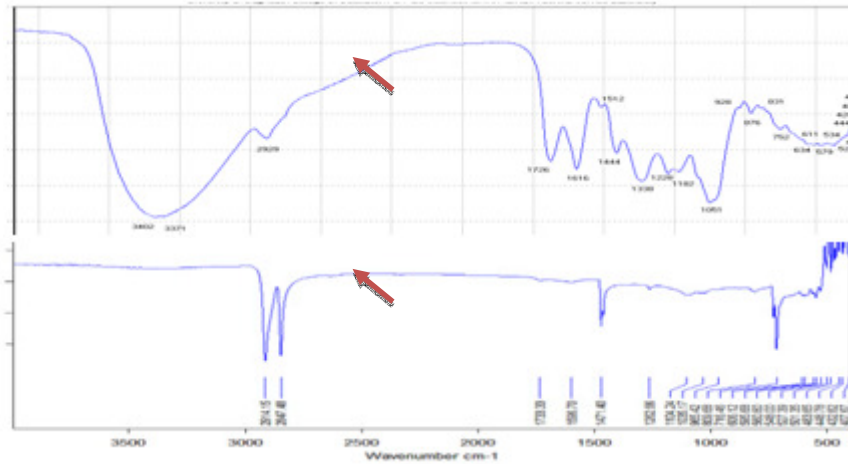


Figure (4) FTIR spectrum where (a): Pomegranate peels powder and (b): UHMWPE Fiber.

Figure (5) shows the FTIR spectrum of polymeric blend nanocomposite specimen [(95%SR (VST-50F): 5%PMMA): 2%PPP], that all the characteristics vibration bands of neat silicone rubbers figure (3) were presented in FTIR spectrum of polymeric blend composite specimen ((SR: 5% PMMA): 2%PPP).

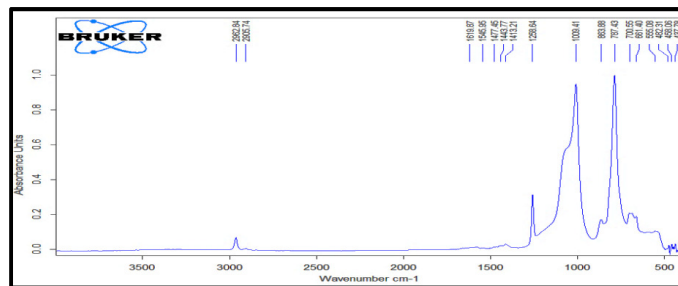


Figure 5 FTIR spectra for polymer blend nanocomposites [(95%SR (VST-50F): 5%PMMA): 2%PPP] reinforced with 2% Pomegranate peels powder.

Figure (6) shows FTIR spectra for hybrid nanocomposite, which is [(SR (VST-50F): 5% PMMA): 0.2% PPP): X% UHMWPE fiber] with different percentages ratios (0.0, 0.5% and 1%) of continuous UHMWPE fibers. These spectra have a very similar to the spectrum of neat silicone rubber (VST- 50F) and spectrum of polymer blend nanocomposites [(95%SR (VST-50F): 5%PMMA): 0.2% (PPP)] and it cannot observe significant differences between these FTIR spectra.

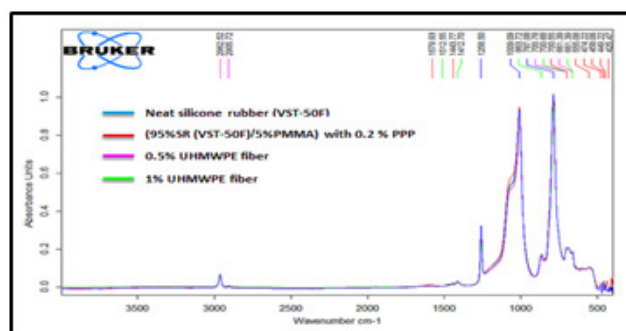


Figure 6 Represent FTIR spectra for silicone rubber (VST-50F) and blend nanocomposite [(95%SR (VST-50F): 5%PMMA): 0.2% Pomegranate peels powder (PPP)] reinforced with 0.5% and 1% UHMWPE Fiber.

4.2. Mechanical Test Results

Figure (7) shows the hybrid nanocomposites [(SR (VST-50F): 5% PMMA): 0.2% PPP): X% UHMWPE] before and after addition of UHMWPE fibers in terms of tensile strength. It can be seen that when adding UHMWPE fibers in polymer nanocomposites, the tensile strength increased. The tensile strength increased from 7.888 MPa for polymer blend polymer blend nanocomposite ((95%SR (VST-50F): 5%PMMA): 0.2% PPP) composite sample to the highest value (8.05 MPa, 10.4 MPa) at 0.5% ratio of both chopped and continues UHMWPE fiber respectively, then decreased to 7.6 MPa and 9.47 MPa at 1% ratio of both chopped and continues fiber reach respectively. This could be attributed to the fact that UHMWPE fibers are described by their high tensile strength and resistance to crack propagation compared with the base polymer nanocomposites; therefore, the hybrid composite specimens can be carried higher load [33-35].

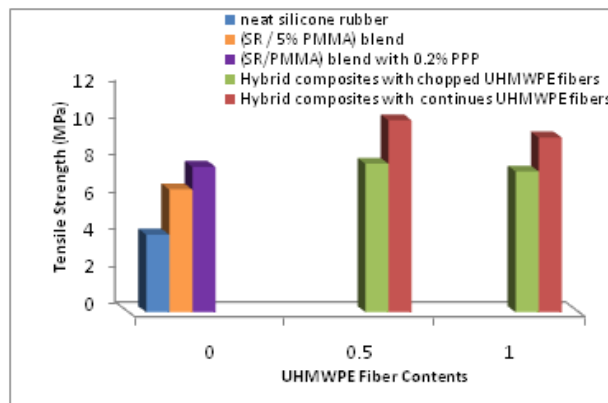


Figure 7 Tensile strength of neat silicon rubber, nanocomposite sample and hybrid composite [(SR (VST-50F): 5%PMMA): 0.2% PPP): X% UHMWPE fiber] verse the fibers amount in specimen.

Figure (8) shows the relationship between the hardness values and the UHMWPE fiber content in polymer blend composite ((SR (VST-50F): 5% PMMA): 0.2% PPP). When adding chopped fibers have less effect on hardness property with increased its content. But adding the continuous UHMWPE fiber gave better hardness property, with increased its percent to 1% of UHMWPE continuous fiber in composite sample to reaches its value to 37 shore A as compared with neat silicone rubber 25shore A and 29 shore A value for the base sample of nanocomposite ((SR (VST-50F): 5% PMMA): 0.2% PPP) [36, 37].

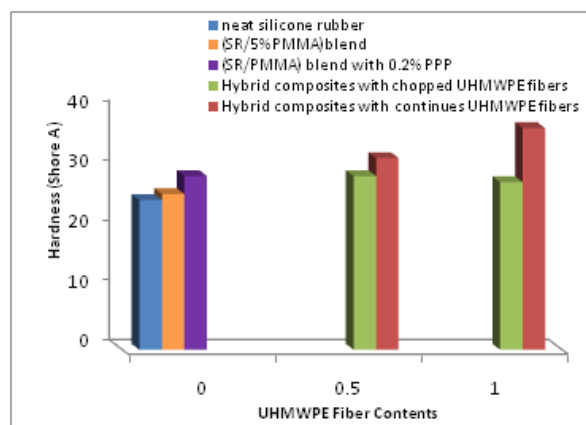


Figure 8 Shore A Hardness of neat silicon rubber, nanocomposite sample and hybrid composite [(SR (VST-50F): 5%PMMA): 0.2% PPP): X% UHMWPE fiber] verse fibers amount in specimen.

Compression set test resulted for the composite specimen ((SR (VST-50F): 5% PMMA): 0.2% PPP) reinforced with UHMWPE fibers are shown in figure (9). It can be noticed that the compression set of hybrid composites increased at the 0.5% of chopped and continuous UHMWPE fibers percent reached to 15.028% and 13.08% respectively, as compared with 12.86% for the base polymer blend composite sample ((SR (VST-50F): 5% PMMA): 0.2% PPP).. But with increased UHMWPE fibers percent to 1% in composite sample, these properties continue to increase reach to the highest value (15.26%, 15.56%) for hybrid composite sample reinforced with both chopped and continuous UHMWPE fibers respectively.

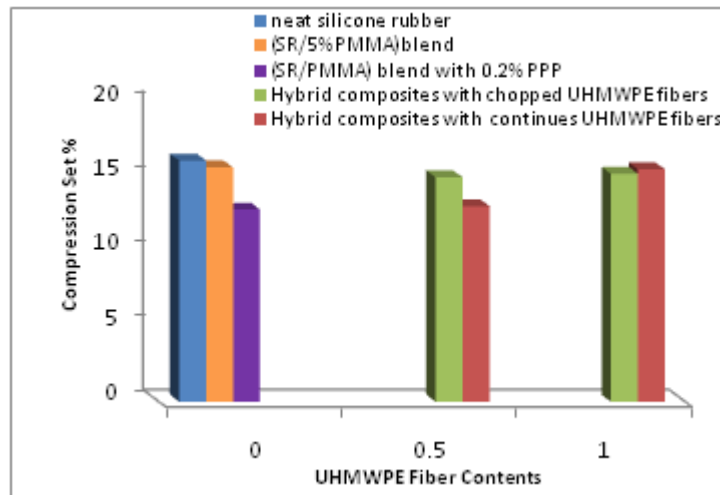


Figure 9: Compression set of neat silicon rubber, nanocomposite sample and hybrid composite [((SR (VST-50F): 5%PMMA): 0.2% PPP): X% UHMWPE fiber] as a function of fibers content in composite.

When hybrid composite samples reinforced with UHMWPE fibers the tear resistance increased with increased fiber content in composite as shown in figure (10), were reached to the optimum values 48N/mm^2 at 0.5% ratio of continuous fiber, but then the tear resistance decline to 34N/mm^2 when the ratio of continuous fiber content increased to 1%, but still higher than the base sample ((SR (VST-50F): 5% PMMA): 0.2% PPP). On the other side, when hybrid composite samples reinforced with chopped UHMWPE fibers, no considerable effect was found in tear resistance property. Based on this result, the increase in tear resistance of nanocomposite when adding UHMWPE fibers to it, may be enhance interfacial surface bonding of fibers and matrix components and that lead to transfer the load from matrix to fibers, this related to the good wettability and good adhesion between the interfaces of these fibers and all constituents of the prepared composites [38].

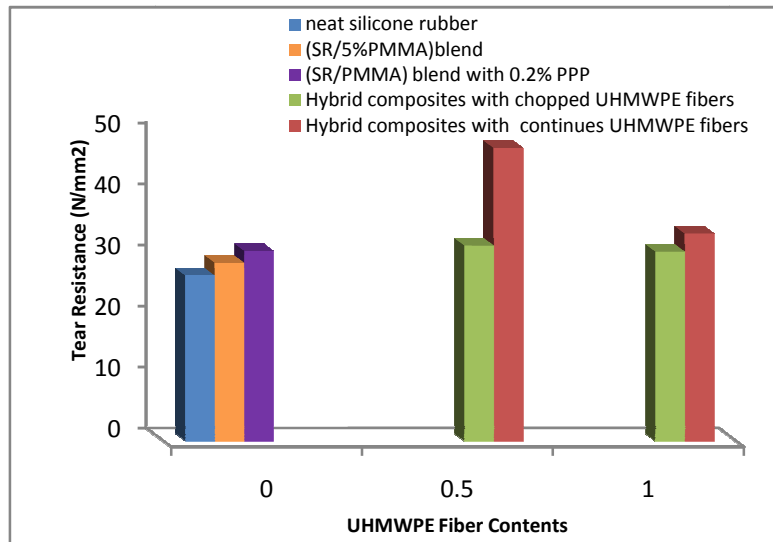


Figure 10 Tear resistance of neat silicon rubber, nanocomposite sample and hybrid composite [(SR (VST-50F): 5%PMMA): 0.2% PPP): X% UHMWPE fiber] as a function of fibers content in composite.

It can be seen from figure(11)that the polymer blend composite sample (SR (VST-50F): 5% PMMA): 0.2% PPP) have surface roughness value equal to (0.128 %).At added chopped UHMWPE fiber to the polymer blend composite sample the surface roughness slightly increased value to (0.129 %, 0.134%) at (0.5% and 1%) respectively percent of chopped UHMWPE fiber, when added 0.5% percent of continuous UHMWPE fiber, the roughness value was increased to 0.13%.

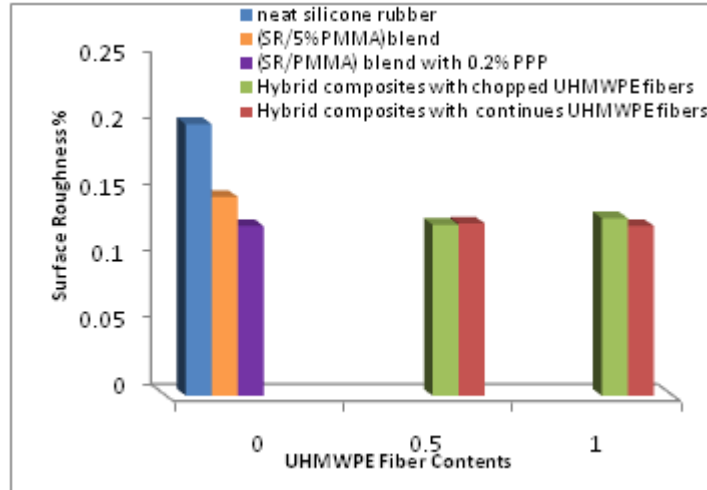


Figure 11 Surface roughness of neat silicon rubber, nanocomposite sample and hybrid composite [(SR (VST-50F): 5%PMMA): 0.2% PPP): X% UHMWPE fiber] as a function of fibers content in composite.

4.3. Density Property Test

Figure(12) shows the relationship between the density of hybrid composite samples and reinforcement fiber content of UHMWPE which were added to sample of polymer blend composite ((SR (VST-50F): 5% PMMA): 0.2% PPP) in both form of chopped and continuous fiber. It was observed from figure 12 that, the density of hybrid composites samples slowly decreased to 1.08 g/cm³ when reinforced with chopped UHMWPE fiber at 0.5% percent,

while the density values remained almost stable for the rest of the hybrid composites samples, either they were reinforced with chopped UHMWPE fiber in other ratio or with continuous fiber in any percentage ratios [39].

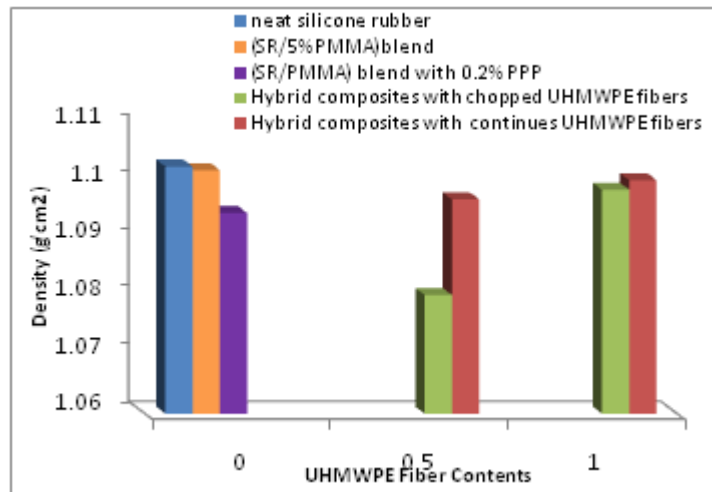


Figure 12 Density property of neat silicon rubber, nanocomposite sample and hybrid composite [(SR (VST-50F): 5%PMMA): 0.2% PPP): X% UHMWPE fiber] verse fibers amount in specimen.

4.4. Water Absorption Results

The relationship between the water absorption of hybrid composites and fiber content UHMWPE in composite, which were added to the base polymeric blend composite ((SR (VST-50F): 5% PMMA): 0.2% PPP) is illustrated in figure (13). It can be observed that the water absorption increases with an increase in the percentage of UHMWPE fibers content in composites. This is due to the affinity of the fibers towards the moisture, and also it may be due to high moisture absorption level of natural fibers in the polymer matrix that results from polar hydroxide groups in the fibers [40-42].

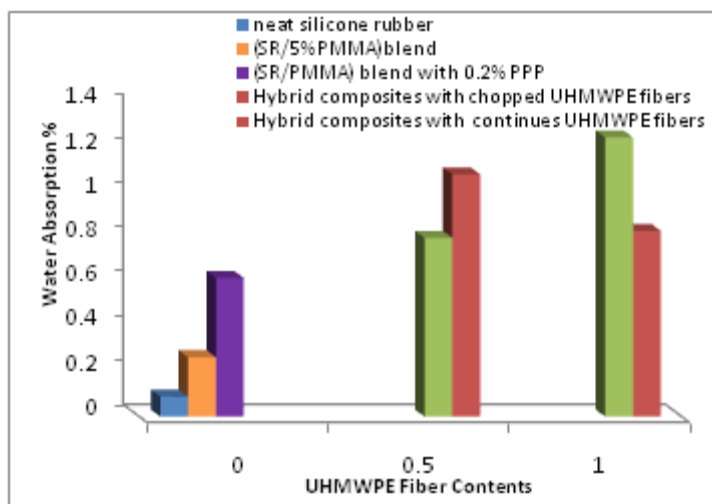


Figure 13 Water absorption of neat silicon rubber, nanocomposite sample and hybrid composite [(SR (VST-50F): 5%PMMA): 0.2% PPP): X% UHMWPE fiber] as a function of fibers content in composite.

4.5. Scanning electron microscopy SEM Test

The photographic imaging for fracture surface morphology of neat silicone rubber and prepared polymer blend ((95% SR (VST-50F): 5% PMMA), are shown in figure (14, a and b) respectively in magnifications ($\times 1000$), which illustrate homogeneous structural morphology and there is no any new phase or phase separated dominants in neat silicone rubber structure. This photographic imaging clearly showed that the polymer blend has a continuous and homogeneous morphology of silicone rubber phase with a random distribution of PMMA phase, which is embedded in matrix material and dispersed as a globule's shapes into the silicon rubber matrix. Furthermore, it can be observed that different sizes of these spherical shapes from PMMA material dispersed randomly in this matrix, which was clearly affect some properties of the prepared polymer blend as compared with neat silicone rubber.

SEM images of the polymer blend nanocomposites with Pomegranate Peels Powder as nanoparticle in composites are shown in figure 14 (c) at magnification ($\times 1000$). It was noticed that most of nanoparticles are embedded inside the matrix material, which act as an integral part of the silicone rubber structure, indicating to better interfacial adhesion between constituents of composite material and good compatibility between the component of silicon rubber blend and the reinforcement nanoparticles, which enhances the mechanical properties [43]. By adding PPP to polymer blend, the morphology of polymer blend nanocomposite does not changes, the structure morphology similar to morphology of polymer blend (figure 14b) and the droplets of PMMA became smaller and more uniform (figure 14 c). During mixing process, the interactions between nanoparticle and polymer blend components mainly occurred through melting of silicone rubber and PMMA molecules, followed by wetting of molten molecules with nanoparticle. After mixing, polymer blend molecule melt encapsulated the surface of nanoparticles and intimate interfacial bonding was formed in nanocomposites as observed from the dense and solid surface especially for nanocomposites when reinforced by 0.2% of pomegranate peels powder [44-46].

Figure (14 d) shows the fracture surface morphology of polymer blend composites ((95% SR (VST-50F): 5% PMMA): 0.2% PPP: 0.5% continuous UHMWPE fiber at magnification 1000x. This morphology shows that most of the UHMWPE fiber has appeared to be embedded in the matrix material, As well as, figure (14) illustrates good uniform distribution of resin with the fabric that leads to good bonding between fibers and matrix. Resin flow and impregnation of the fibers can be observed in the SEM micrographs shown through the red arrows in figure 14 d, this suggests that this polymer blend composite sample has been efficient in holding on to the fibers. This indicates that higher mechanical strength the composites specimen have high degree of fiber orientation, which lead to higher mechanical strength [47].

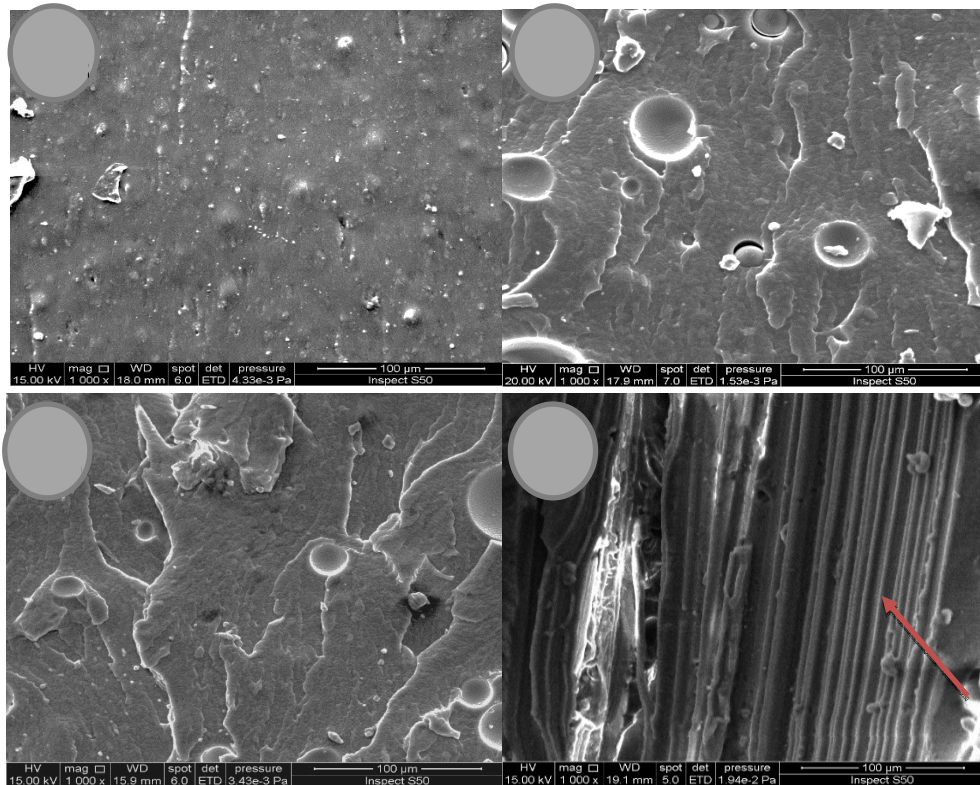


Figure (14) SEM Image of fracture Surface Morphology for (a and b): neat silicone rubber and polymer blend (95% SR (VST-50F): 5% PMMA) each of them at magnification (x1000), (c and d): for polymer blend composites ((95% SR (VST-50F): 5% PMMA): 0.2% Nano Pomegranate peels powder and polymer blend hybrid composites ((95% SR (VST-50F): 5% PMMA): 0.2% PPP: 0.5% continuous UHMWPE fiber each of them at magnification.

5. CONCLUSIONS

The hybrid composites samples were prepared by adding UHMWPE fiber to the sample of polymeric blend (95% SR (VST-50F): 5% PMMA) composites which reinforced with 0.2% of pomegranate peels powder. The results showed the following items:-

- From FTIR spectrum there are no other new peaks were observed for hybrid composites samples, which reinforced by continuous UHMWPE fiber compared with neat silicon rubber and polymeric blends of SR/PMMA specimens with the addition of 0.2% Nano Pomegranate peels powder.
- A hybrid composites samples, which reinforced by 0.5% of continuous UHMWPE fiber having the highest values of tensile strength and tear resistance which reached to 10.4 MPa, 48 N/mm² respectively, a hybrid composites samples, which reinforced by continuous UHMWPE fiber having lower values of the surface roughness as compared with other sample reinforced by chopped UHMWPE fiber. A homogeneous structural morphology was observed in SEM test, this indicated to good interactions between the constituents of all polymer blend composites samples.
- The hybrid composites under the stress load have a high degree of fiber orientation, which leads to higher mechanical strength. Therefore, these samples may be a achieve the most properties of maxillofacial applications.

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