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Effect of Accelerated Weathering on the Compressive Strength for PMMA Nano Composites and PMMA Hybrids Nano Composites Used in Dental Applications

Abstract- *In the present research, efforts are made to develop the properties of PMMA resin that used for upper and lower prosthesis complete denture, by addition four different types of nanoparticles powders, which are fly ash, fly dust, zirconia and aluminum that added with different ratios of volume fractions of (0.01, 0.02 and 0.03) to poly methyl methacrylate (PMMA), cold cured resin (castavaria) is the new fluid resin (pour type) as a matrix. The nano composite and hybrid nano composite for prosthetic dentures specimens, preparation was done by using (Hand Lay-Up) method as six groups which includes: the first three groups consists of PMMA resin reinforced by fly ash , fly dust and ZrO2 nanoparticles respectively, the second three groups consists of three types of hybrid nano composites, which includes ((PMMA:X% fly ash) - (1%Al + 3%ZrO2)), ((PMMA:X% fly dust) - (1% Al + 3%ZrO2)) and ((PMMA:nZrO2) - (1% fly ash+ 3% fly dust)) respectively. As well as, the effect of moisture and UV was taking into consideration in this study. The compression test results shows that the values of compressive strength, compressive elastic modulus, and compressive strength under the effect of accelerated weathering (moisture and UV radiation) increased with the addition of nano powders (fly ash, fly dust, zirconia and aluminum). As well as, the results showed that the maximum values of compressive strength reach to (286.25MPa) for (PMMA + 2%nZrO2) nano composite. In addition, the results showed that the compressive elastic modulus reach to the maximum value (25.4166GPa) in the nano composite material (PMMA + 2%nZrO2). Moreover, the results showed that the compressive strength under the effect of accelerated weathering (moisture and UV radiation) reach to the maximum value to (315MPa) for the nano composite material (PMMA + 3%nZrO2).*

Keywords- *Hybrid Nano Composites, PMMA, Fly Ash Nanoparticles, Fly Dust, Aluminum, Zirconium Oxide, Compressive Strength, Accelerated Weathering.*

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1. Introduction

A biomaterial can be defined as any synthetic or natural material that is used to replace or restore function to a body tissue. Complete or partial denture base material represented one type of biomaterials that must be have good stiffness, toughness, hardness and resistance to wear and abrasion, and good thermal conductivity, thermal diffusivity, and dimensional stability, non-toxic or allergic, color stability, good chemical stability, in addition to esthetical pleasing and use in the oral cavity [1]. This material is still not enough to achieve the ideal mechanical requirements for dental applications although it is the most commonly used in dentistry for fabrication of denture bases. This problem was attributed mainly to its low plaque accumulation and low fracture resistance [2-3]. Composites are multiphase materials that are chemically dissimilar and artificially made and separated by distinct interface [4]. Particulate composite materials consisting of polymer resin as matrix and particles as reinforcement phase. The particles carry a major portion of the load. The particles are used to increase the modulus and decrease the ductility of the matrix. Polymer composite materials reinforced with particles (ceramic, metal particles) can be used for various engineering applications to provide unique mechanical and physical properties with a low specific weight. In order to achieve better mechanical strength, it is usually reinforced with ceramic powders or fibers (aramid, carbon and glass). Ceramic particles with small size are known to enhance the tribological and mechanical properties of polymers [5]. Fly ash, an industrial waste, because it is a mixture of

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oxide ceramics, it can be used as a potential filler material in polymer matrix composites It improves the mechanical and physical properties of the composites [6]. Some researches, which are accomplished in this field, it's:

Sood, and Schajpal added powders such as aluminum (99.9 %) pure , copper or silver with average particle size of 10 micro meter into PMMA acrylic denture base material resin in different volume fraction of (5%, 10%, 15%, 20% and 25%). The addition of these metal fillers showed increases in compressive strength and decreases in tensile strength as the percentage of metal fillers increases. Thermal conductivity increased progressively with the addition of metal fillers but did not proportionally, as the metal fillers volume fraction increased [7].

Tham et al, investigated the effects of Simulated Body Fluid (SBF) and water absorption on the flexural properties of PMMA/HA composites for an immersion duration of 2 months. In order to enhance the interfacial interaction between the PMMA and HA, silane-coupling agent [3-methacryloxypropyltrimethoxy silane (γ-MPS)] was used. It was found that flexural strength of the PMMA/HA composites after SBF absorption and water absorption was decreased due to the plasticizing effect of water molecules [8].

Shyang studied the effect of the addition of hydroxyapatite (HA) particles on the flexural properties of a heat polymerizing PMMA denture base resin. The results showed that the flexural modulus, flexural strain and flexural strength of PMMA/HA composites were decreased with the addition of hydroxyapatite (HA) particles [9].

Khalaf investigated the effect of the addition of siwak micro powder on the Certain Mechanical Properties of Acrylic Resin. The results showed that the addition of (7 %) siwak powder to the Acrylic Resin revealed a significant decrease in compressive strength, impact strength and tensile strength [10]. The one recent study mentioned elsewhere, which involved the numerical study by the tensile properties analysis of the prosthetic dentures, which prepared from the same of composite material maintained in the reference above, and the numerical analysis results of the finite element method shown the some agreement with the experimental results [11].

Salih et al. investigated the effect of the addition of nano hydroxyapatite (nHA) particles, micro zirconia (ZrO_2) particles on the fatigue strength and compression strength of the composite prosthetic denture. The compression test result shows that the values of compression strength increased with increasing the volume fraction of (nHA and ZrO_2) particles for all groups' specimens. In addition, the results showed the (PMMA-ZrO_2) composite has greater values for compression strength. As well as the results shows that the maximum value of compression strength for hybrid laminated composite is obtained in hybrid laminated composite materials (PMMA- ZrO_2)-5% Glass Fiber [12].

Another study about the flexural properties and impact strength for PMMA prosthetic complete denture base reinforced with hydroxyapatite nano particles and with zirconia micro particles. The results study showed the values of most properties increase with increasing of the volume fraction of hydroxyapatite and ZrO_2 particles in polymer composite materials, while, the impact strength decreased [13].

The objective of the current work is attempts to develop a PMMA resin, which is used in the denture base and in dental prosthesis applications. Through study the effect of adding different nanoparticles powders, on the compression properties for PMMA nano composites, and hybrids nano composites. Moreover, study the effect of the UV radiation and moisture in sequentially on the compressive strength for the nano PMMA composites, as well as the hybrid nano composites, which use for the prosthetic denture.

2. Materials and Methods

I. Materials Used

In this research poly methyl methacrylate (PMMA) cold curing as new pour (fluid) resin type (Castavaria) has been used, provided from (Vertex – Dental Company). Table 1 shows some of the mechanical and physical properties of cold cure PMMA according to the supplied company. Four types of nanoparticles powders were used as reinforces materials with selection volume fraction of (0.01, 0.02 and 0.03) including: the fly ash nanoparticles (nF.A) class B obtained from the England with dark gray color, fly dust nanoparticles (nF.D) obtained from the cement plants in Kufa with Yellowish brown color. Table 2 and Table 3 shows the chemical composition analyses of fly ash and fly dust nanoparticle powders respectively which was used in this research, zirconium oxide nanoparticles (nZrO_2) were supplied as partially stabilized particles form, which provided from (ZIRCON Company in England) and aluminum nanoparticles with dull gray color.

Table 1: Some Mechanical and physical properties of neat pmma resin used in this research according to the company processed (vertex–dental company)

Young's Modulus (GPa)	Impact Resistance (KJ/m^2)	Flexural Strength (MPa)	Flexural Modulus (GPa)	Water Sorption (µg/mm^3)	Solubility (µg/mm^3)	Water Absorption (%)	Density (gm/cm^)
1.63-3	8.3	79	2.3	23.2	1.8	2.5	1.19

Table 2: Chemical composition analyses of fly ash used in this research

Element Oxide	$SiO_2Al_2O_3$	TiO_2	MgO	K_2O	CaO	Fe_2O_3	Mn_2O_3	Na_2O	P_2O_3	L.O.I
The weight (%)	58.227.7	1.4	0.05	3.59	0.84	4.99	0.31	0.74	0.34	1.84

Table 3: Chemical composition analyses of fly dust used in this research

Element Oxide	SiO_2	Al_2O_3	LiO	MgO	CaO	Fe_2O_3	L.O.I
The weight (%)	12.30	3.02	29.30	4.80	38.08	2.91	9.48

II. Preparation Methods and Curing Cycle of Test Specimens

The PMMA nano composite materials and hybrid nano composite materials specimens were prepared by using the Vertex™-Castavaria. According to the manufacturer's instructions of Manufacturer Company, the standard proportion in mixing ratio for cold cure PMMA resin is (1 ml) (0.95g) monomer liquid (MMA) and (1.7 g) acrylic powder (PMMA).

The Vertex acrylic Castavaria is moldable, where the liquid monomer (MMA) was placed in dry glass container, followed after that with slow addition of dry powder (PMMA) to the liquid monomer (MMA). After pouring completion into the metallic mould, the metallic mould was placed in the multi cure system (Ivo met) manufactured by Vertex-dental company according to the polymerization curing instructions at temperature equal to (55°C) and pressure equal to (2.5 bar) for (30 min) in order to complete the polymerization process of the acrylic specimens.

After the polymerization curing completed, the specimens were de molding to remove from the metallic mould with very smooth upper and lower surface.

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III. Composites and Hybrid Composites Specimens.

Six groups of specimens which prepared in this research for the prosthetic denture base, includes, the first three groups, is prepared as a nano composite specimens which divided into nine nano composites, consists of PMMA resin reinforced by fly ash, fly dust and ZrO_2 Nanoparticles respectively, and the second three of groups, divided into nine specimens consists of three groups of hybrid nano composites, which are ((PMMA: X% fly ash) - (0.01 Al + 0.03 ZrO_2)), ((PMMA: X% fly dust) - (0.01 Al + 0.03 ZrO_2)) and ((PMMA : nZrO_2) - (0.01 fly ash + 0.03 fly dust)) respectively.

According to the concentration of the reinforcement, materials for all specimens of these groups are shown in the Table 4.

IV. Atomic Force Microscopic (AFM)

Test Atomic Force Microscope is used to measuring the average particle size of the nano powders materials, which is shown that the average diameter for each of fly ash, fly dust, ZrO_2 and aluminum are (64.94nm), (84.23nm), (84.35nm) and (53.87nm) respectively. The results of particle size distribution for these nano powders is shown in the Figure (1 (a, b, c and d)) respectively.

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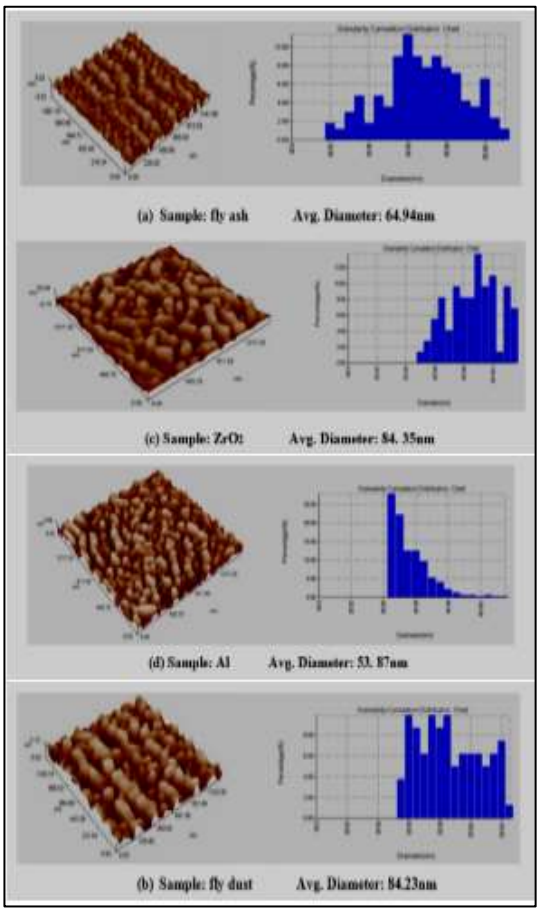

Figure 1: Atomic force microscopy test for nano powders (a) fly ash (b) fly dust(c) zirconium oxide and (d) aluminum

3. Mechanical Testing

In order to evaluate the compression properties of the PMMA nano composite materials and PMMA hybrid nano composite materials of the denture prosthetic materials, compression test was performed in this research. According to the (ASTM D695) the compression test was performed at room temperature by using the universal tensile test machine manufactured by (Laryee Company in china), type is (WDW-50). The crosshead speed was (0.2mm/min) and the load was applied gradually until the fracture of the specimen occurs [14]. According to (ADA Specification No.12, 1999), all the test specimens after preparation and polishing processes must be stored in distilled water at (37± 1°C) for 48 hr [15].

4. Weathering Testing

In order to evaluate the compressive strength under the effect of accelerated weathering (moisture and UV radiation) of the PMMA nano composite materials and PMMA hybrid nano composite materials of the denture prosthetic materials, accelerated weathering (QUV) test were performed in this research. The accelerated weathering test was performed according to the (ISO 4892-3) by using QUV accelerated weathering test device, model is (QUV/spray), manufactured by (Q-labs in Australian), type is (UV-A340 lamps) [16]. The fluorescent UV devices offer a condensation cycle during lights-off period to produce moisture; therefore, the test specimen surface is exposed to heated, saturated moisture of air and water vapor [17]. It is possible to simulate 63 years of natural UV radiation exposure on a test device in a single year. The temperature inside the device was equal to 55°C and the irradiance (UV) incident on the specimens was 0.58(W/m^2). The total time for testing was 120 hr included subjecting the specimens to 2 hr UV and 2 hr moisture [18].

Table 4: PMMA nano composite specimens and hybrid nano composite specimens that prepared in this research

Material	
Cold Cure Pure PMMA as Reference Material	
Nano Composite Number	Matrix+%Volume Fraction of NanoParticles
Nano Composite 1	PMMA+0.01 nano fly ash
Nano Composite 2	PMMA+0.02 nano fly ash
Nano Composite 3	PMMA+0.03 nano fly ash
Nano Composite 4	PMMA+0.01 nano fly dust
Nano Composite 5	PMMA+0.02 nano fly dust
Nano Composite 6	PMMA+0.03 nano fly dust
Nano Composite 7	PMMA+0.01 $nZrO_2$
Nano Composite 8	PMMA+0.02 $nZrO_2$
Nano Composite 9	PMMA+0.03 $nZrO_2$
Hybrid Nano Composite Number	(Matrix) +%Volume Fraction of nanoParticle
Hybrid Nano Composite 1	(PMMA+0.01 nano fly ash) + (0.01Al and 0.03 ZrO_2)
Hybrid Nano Composite 2	PMMA+0.02 nano fly ash) + (0.01Al and 0.03 ZrO_2)(
Hybrid Nano Composite 3	PMMA+0.03 nano fly ash) + (0.01Al and 0.03 ZrO_2)(
Hybrid Nano Composite 4	PMMA+0.01 nano fly dust) + (0.01Al and 0.03 ZrO_2)(

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Hybrid Nano Composite 5	PMMA+0.02 nano fly dust) + (0.01Al and 0.03 ZrO_2)(
Hybrid Nano Composite 6	PMMA+0.03 nano fly dust) + (0.01Al and 0.03 ZrO_2)(
Hybrid Nano Composite 7	PMMA +0.01 nZrO2) + (0.01fly ash and 0.03 fly dust)(
Hybrid Nano Composite 8	PMMA+0.02 nZrO2) + (0.01fly ash and 0.03 fly dust)(
Hybrid Nano Composite 9	(PMMA+0.03 nZrO2) + (0.01fly ash and 0.03 fly dust)

5. Results and Discussion

The compressive strength and compressive elastic modulus values of neat PMMA resin, PMMA nano composites and PMMA hybrid nano composites for all samples that were prepared in this research before exposure to accelerated weathering (UV radiation and moisture in sequentially) are presented in the Figures 2-9.

Figure 2 and Figure 3 show the effect of adding various types of nanoparticles powders (fly ash, fly dust and zirconium oxide) on the compressive strength and compressive elastic modulus for PMMA nano composites respectively. It can be noted from these figures that the addition of the fly ash, fly dust with volume fraction (0.01 and 0.02) and zirconium oxide with volume fraction (0.01, 0.02 and 0.03) lead to increase the compressive strength and compressive elastic modulus of the PMMA nano composites and reach to maximum value at (0.02) of volume fraction as comparing with neat PMMA. This is due to the high interfacial shear strength between the PMMA matrix and nanoparticles and this leads to the formation of strong physical bonds, which in turn prevent the propagation of the cracks inside the material. As well as, the propagation of the crack can be changed by good bonding between the PMMA matrix and nanoparticles [19 and 20]. Moreover, the incorporation of the hard nanoparticles powders into the polymer matrix improves the stiffness of the composites by restricted the mobility of the matrix chains [21]. As well as, good distribution of nanoparticles powders in composite material, especially at the low concentrations of nanoparticles additives to the composite as shown in figure 4, and this it will may be reduced agglomeration of the nanoparticles, and that may be lead to reduces stress concentration in composite material near the agglomerated nanoparticles, so, such small stresses are not sufficient enough to break the weak interactions at the interface [22]. Therefore, these small stresses can be easily transferred from the matrix to the nanoparticles, so allowing the particles to contribute its high brittleness property to the nano composites so, the compressive strength and compressive elastic modulus increases [23]. Overtime, the formation of a strong structure of the PMMA nano composite materials which depending on the formation of strong interfaces bonding between the reinforcing nanoparticles and PMMA matrix, so that, the resultant is nano composite materials with strong physical bonding, therefore required high compressive stress to break it, and this lead to increasing compressive strength and compressive elastic modulus [24]. On the contrary, it can be noted from the Figure 3 that the addition of the fly ash and fly dust nanoparticles to larger than (0.02) volume fraction leads to decrease the compressive elastic modulus of the PMMA nano composites. This is due to the high volume fractions of the nanoparticle powders leads to agglomeration of these nano powders together; therefore, these powders play an important role in stress concentration. Therefore, when the compressive stress was applied on the specimen, the value of the stress concentration increases dramatically near the agglomerated nanoparticles and making the debonding between PMMA and nano powders and this cause cracks propagate faster inside the material so that, the fracture occurs immediately [25 and 26]. Further to that a bad wettability between the nano particles and matrix, especially at high concentrations, so that, the resultant is nano composite material with weak physical bonding, and this required low compressive stress to break the sample [24]. It is worth mentioning, although there is a change in the type and proportions of the components of each of the fly ash and fly dust, which is showed earlier through the Tables 3 and 4, but the effect of each of these components on the compressive elastic modulus for the prepared nano composite samples it was symmetric at volume fractions (0.01 and 0.02).

In addition, it is observed from Figures 2 and 3 that the nano composites materials reinforced with the zirconium oxide nanoparticles have the higher values of compressive strength and compressive elastic modulus, as compared with their counter parts of the other groups of the nano composites materials, which reinforced with fly ash and fly dust nanoparticles. The reasons behind such a behavior are that the compressive strength of the zirconium oxide higher than the compressive strength of the fly ash and fly dust, as well as, have good compatibility between constituents of composite materials [24].

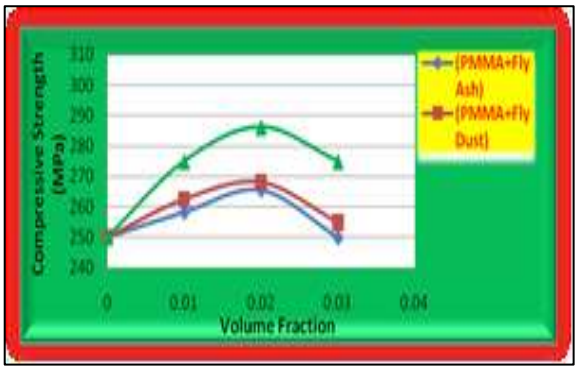


Figure 2: Compressive strength of PMMA nano composite materials as a function of volume fraction of nanoparticles (fly ash, fly dust and ZrO2) in PMMA matrix

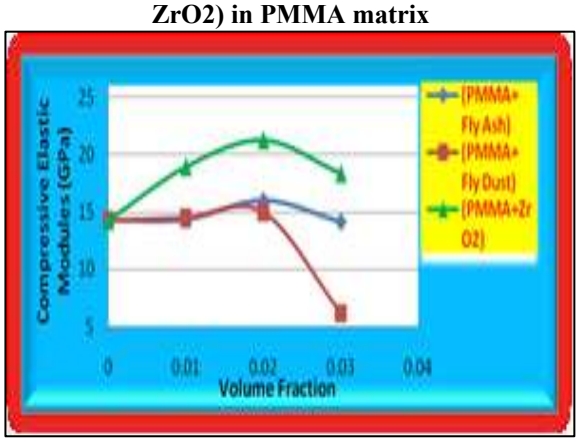


Figure 3: Compressive elastic modulus of PMMA nano composite materials as a function of volume fraction of nano particles (fly ash, fly dust and ZrO_2) in PMMA matrix

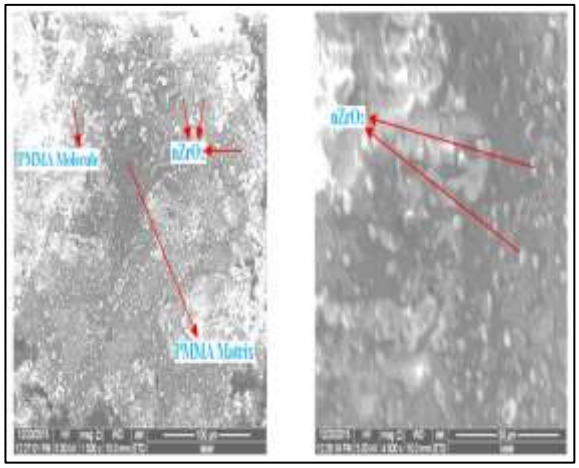


Figure 4: SEM images showing the surface morphology for nano composite (PMMA = 0.01 ZrO_2) at magnifications (1000× and 4000×)

The effect of the addition of the mixture of nano particles powders (0.01 Al and 0.03 ZrO_2) to the nano composite materials (PMMA:X% fly ash) and (PMMA:X% fly dust), on the compressive strength and compressive elastic modulus for these hybrid Nano composite materials ((PMMA: X%fly ash) + (0.01 Al and 0.03 ZrO_2)) and ((PMMA: X%fly dust) + (0.01 Al and 0.03 ZrO_2)), it was shown in Figures (5-8). It was noticed that the addition of the mixture of nano particles powders with ratio of (0.01 Al and 0.03 ZrO_2) to the nano composite materials (PMMA:X% fly ash) and (PMMA:X% fly dust) respectively, lead to increase in the compressive strength and compressive elastic modulus of the hybrid nano composites as comparing with their counterparts of nano composites which are (PMMA:X% fly ash) and (PMMA:X% fly dust) of the same volume fraction of fly ash and fly dust respectively. This behavior was related to the addition of the zirconium oxide that have compressive strength higher than the compressive strength of the fly ash and fly dust as previously mentioned, in addition to the improvement of the mechanical properties that is associated with the addition of zirconium oxide nanoparticles [24]. On the contrary, it can be noted in Figure (6) that the addition of the mixture of nano particles powders with ratio of (0.01 Al + 0.03 ZrO2) to the nano composite materials (PMMA:0.03 fly ash) leads to decrease in the compressive elastic modulus of the hybrid nano composite as comparing with its counterpart of nano composite which is (PMMA:0.03 fly ash). This is due to the agglomeration effect, bad wettability and weak physical bonding at high volume fractions of the mixture of hybrid nanoparticles powders which are mentioned in above item for Figures (2 and 3) [24, 25 and 26].

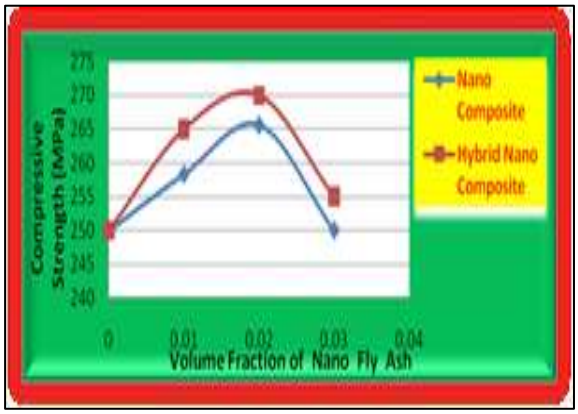
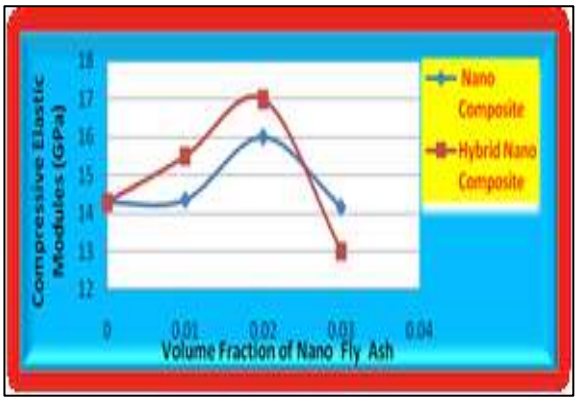


Figure 5: Compressive strength for PMMA nano composite and PMMA hybrid Nano composite materials as a function of nano fly ash in PMMA matrix



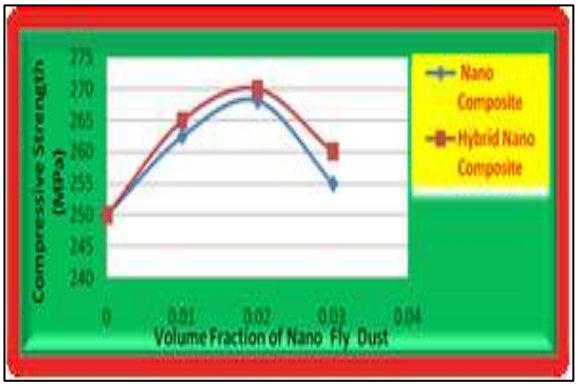


Figure 7: Compressive strength for PMMA Nano composite and PMMA hybrid Nano composite materials as a function of nano fly dust in PMMA matrix

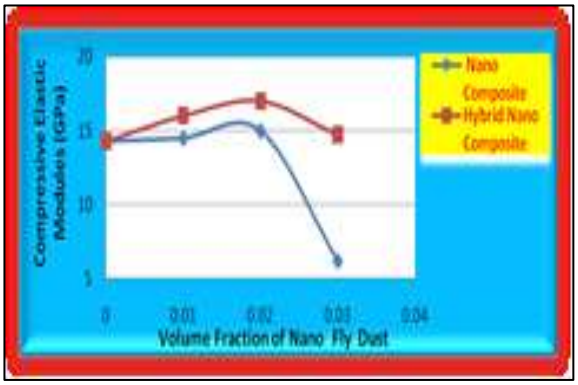


Figure 8: Compressive elastic modulus for PMMA nano composite and PMMA hybrid nano composite materials as a function of nano fly dust in PMMA matrix

On the contrary, it can be noted from Figures 9 and 10 that the addition of the mixture of nanoparticles powders with ratio of (0.01 fly ash + 0.03 fly dust) to the nano composite materials (PMMA: X%ZrO_2) lead to decrease in the compressive strength and compressive elastic modulus of the hybrid nano composites (PMMA: X%ZrO_2) + (0.01 fly ash + 0.03 fly dust) as comparing with their counterparts of nano composites which is (PMMA: X%ZrO_2) of the same volume fraction of ZrO_2. The reasons are that the high concentrations of the nanoparticle such as fly ash and especially with fly dust leads

Figure 6: Compressive elastic modulus for PMMA nano composite and PMMA hybrid nano composite materials as a function of nano fly ash in PMMA matrix

to agglomeration and stick of these nano powders together, therefore these agglomerated powders play an important role in stress concentration which occurs near the agglomerated powders, and this cause cracks propagate faster inside the material so that, the fracture occurs immediately as mentioned in the previous item for Figures 2 and 3 [25 and 26].

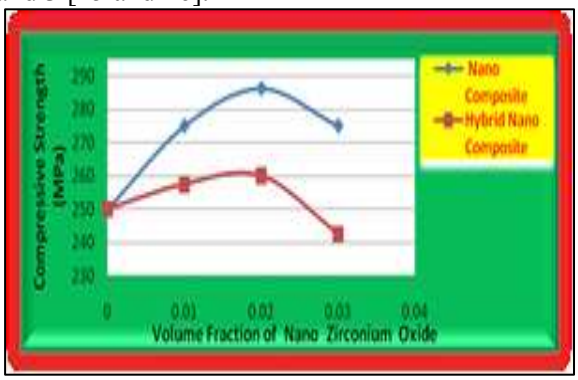


Figure 9: Compressive strength for PMMA nano composite and PMMA hybrid nano composite materials as a function of nano zirconium oxide in PMMA matrix

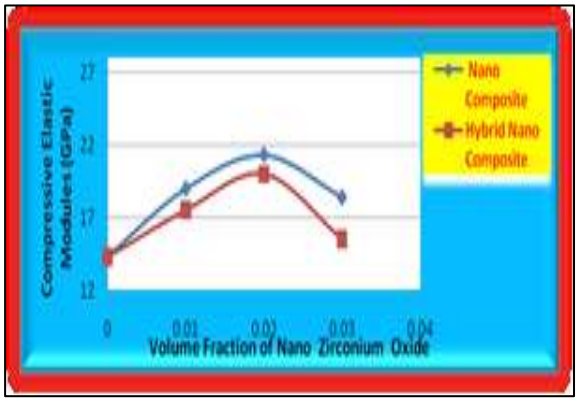


Figure 10: Compressive elastic modulus for PMMA nano composite and PMMA hybrid nano composite naterials as a function of nano zirconium oxide in PMMA matrix

The compressive strength values of neat PMMA, PMMA nano composite and PMMA hybrid nano composite materials for all samples that were fabricated in this research after and before exposure to accelerated weathering (UV radiation

and moisture in sequentially) are presented in the figures 11-16.The main purpose of this test is to select which volume fraction (0.01, 0.02 or 0.03) of nanoparticles powders will stabilize the PMMA nano composite and PMMA hybrid nano composite materials under the effect of concentrated UV radiation and humidity. From these Figures it is found that the compressive strength values of the neat PMMA and PMMA nano composites after exposure to accelerated weathering (UV radiation and moisture) higher than their values before exposure to accelerated weathering (UV radiation and moisture in sequentially), this is due to the concentrated UV thermal energy leads to completion of the polymerization process which leads to formation strong secondary bounds (cross linking) between the carbons chains, less residual monomer and less porosity [24]. On the other hand, Figure 11 and Figure 12 show the effect of adding various types of nanoparticles (fly ash and fly dust) on the compressive strength for PMMA nano composites after and before exposure to UV radiation and humidity in sequentially. It can be noted from these Figures that there is a noticeable effect of accelerated weathering on the compressive strength. It was found there is an increase in values of compressive strength of the PMMA nano composite materials as compared with their counterparts, of the other specimens before exposure to UV radiation and moisture in sequentially. Moreover, it has been observed that there is a symmetrical behavior in the values of compressive strength of PMMA nano composites when adding nanoparticles to it either before or after exposure to accelerated weathering.

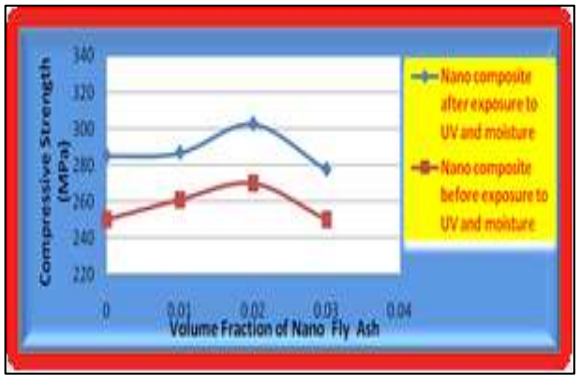


Figure 11: Compressive strength of PMMA nano composite materials either before or after exposure to accelerated weathering as a function of volume fraction of fly ash nano particles in PMMA matrix

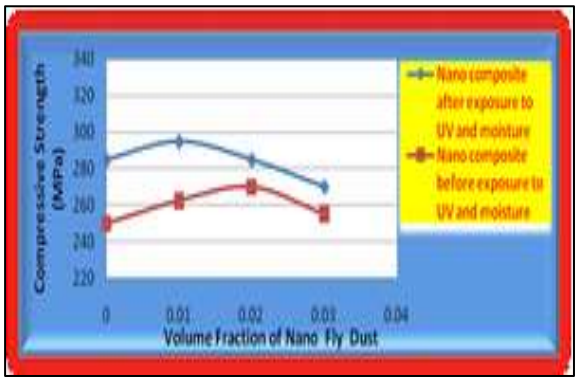


Figure 12: Compressive strength of PMMA nano composite materials either before or after exposure to accelerated weathering as a function of fly ash nanoparticles in PMMA

Figure 13 shows the effect of adding zirconium oxide nanoparticles on the compressive strength for PMMA nano composites after and before exposure to UV radiation and humidity in sequentially. It can be noted from this Figure that there is a noticeable effect of accelerated weathering on the compressive strength. In addition, it was noticed that there is an increase in values of compressive strength for the PMMA nano composite with an increase in the ratio of zirconium oxide in nano composite. Furthermore, this behavior is asymmetrical with their counterparts of the samples that tested before exposure the samples to UV radiation and humidity. Nevertheless, the compressive strength values were decreased when adding ZrO_2 at volume fraction 0.03 in nano composite. reasons behind such a behavior for Figures 11-13 are that the addition of the nanoparticle such as fly ash, fly dust and especially zirconium oxide prevent the degradation of the PMMA nano composite materials matrix by ultraviolet light by preventing the radicalization degradation of an ester side group which leading to prevent β-scission of the polymer carbon chains backbone [27]. Moreover, the addition of these nanoparticles lead to decreasing moisture absorption because of the fact that these nanoparticles replace hydrophilic PMMA resin for the nano composite materials, result in a decrease in moisture absorption since diffusivity of water molecules through these materials is greatly lower than that through the PMMA matrix. In addition, strong interfacial bonding between PMMA polymer and constituents of nanoparticles (fly ash, fly dust and zirconium oxide) lead to decreases micro voids in the PMMA nano composite materials, and because of this decreasing of moisture absorption, which lead to minimum volumetric expansion between the PMMA resin and the nano fillers, therefore, the thermal stresses generated by the

UV radiation, not exceeds the strength of the interphase region between the PMMA resin and these nano fillers. Therefore, de bonding not takes place between the nano fillers and the PMMA resin matrix resulting in an increasing in the compressive strength of the dental PMMA nano composite materials [28]. Moreover, the increase in the compressive strength also can be attributed to the concentrated energy of UV radiation, which leads to completion of the polymerization process of residual monomer in PMMA matrix, which leads to formation of strong physical bonds (cross linking) between the PMMA carbons chains, and then less residual monomer and less the porosity, which adversely influence the compressive strength. In addition, the increase in the compressive strength can be also attributed to the increasing in the interfacial shear strength (adhesion bonding) between the PMMA matrix and nanoparticles due to the absorbed moisture. So that, the resultant is nano composite materials with strong physical bonding required high compressive stress to break it. Based on the foregoing, the compressive strength values of PMMA nano composites (with the volume fraction no more than 0.02) after exposure to UV radiation and moisture higher than the compressive strength values of their counterparts of the PMMA nano composites (with the volume fraction no more than 0.02) before exposure to UV radiation and moisture [24]. On the contrary, it can be noted from figures 11 and 12 that the addition of the fly ash and fly dust nanoparticles size with volume fraction more than 0.02 leads to decrease in the compressive strength of the PMMA nano composites after exposure to UV radiation and moisture in sequentially as comparing with net PMMA. The reasons behind such a behavior are that at volume fraction more than 0.02, the degradation of the PMMA nano composites materials by ultraviolet light is occur due to the high energy incident radiation initiates radicalization of an ester side group leading to β-scission of the polymer backbone and decomposition of the radical end to products including H_2, CO, $COOH_2$, CH_2 and HCOOCH [27]. The loss of the ester group creates a greater free volume (voids) in the PMMA matrix, and as a result of this, increasing of moisture absorption which lead to sufficient volumetric expansion between the PMMA matrix and the nano fillers, therefore, the thermal stresses generated by the UV radiation exceeds the strength of the interphase region between the PMMA matrix and the nano fillers. So, debonding takes place between the nano fillers and the PMMA matrix resulting in decreasing in the compressive strength of the dental PMMA nano composite materials [28]. The effect of the addition of the mixture of nanoparticles powders (0.01 Al and 0.03 ZrO2) to nano composites (PMMA: X% fly ash) and (PMMA: X% fly dust), as well as the addition of the mixture of nanoparticles powders (0.01 fly ash+0.03 fly dust) to nano composites (PMMA: X%nZrO2) , on the compressive strength for these hybrid nano composite materials ((PMMA: X% fly ash) + ((0.01 Al and 0.03 ZrO2)), ((PMMA: X%fly dust)+(0.01 Al and 0.03 ZrO2)) and ((PMMA: X%n ZrO2)+(0.01 fly ash and 0.03 fly dust)) after and before exposure to UV radiation and moisture in sequentially, it was shown in Figures 14-16. It can be noted from these Figures that there is a noticeable effect of accelerated weathering on the compressive strength behavior of the PMMA hybrid nano composite materials after exposure to UV radiation and moisture in sequentially, as comparing with their counterparts of the other specimens of the PMMA hybrid nano composite materials before exposure to UV radiation and moisture. From Figures 14 and 15 show that the compressive strength values for the neat PMMA resin and hybrid nano composite materials ((PMMA: 0.01 and 0.02 fly ash) + (0.01 Al + 0.03 ZrO2)) and ((PMMA: 0.01 and 0.02 $nZrO_2$) – (0.01 fly ash + 0.03 fly dust)) after exposure to UV radiation and moisture is higher than the compressive strength values of their counterparts of the pure PMMA, PMMA hybrid nano composites of the same volume fractions before exposure to UV radiation and moisture. The reasons behind such a behavior are related to the same reasons, which mentioned in the previous item for Figures 12 and 13. Furthermore, it can be noted in Figure 14 that the addition of the mixture of nano particles with ratio of (0.01 Al + 0.03 ZrO2) to the Nano composite material (PMMA: 0.02 fly ash) as a matrix lead to increase in the compressive strength of the PMMA hybrid nano composite ((PMMA: 0.02 fly ash) - (0.01 Al + 0.03 ZrO2) after exposure to UV radiation and moisture as comparing with net PMMA. The reasons behind such a behavior are related to the same reasons, which mentioned in the previous item for Figures 12 and 13. Moreover, it can be noted in Figures (14-16) that the addition of the mixture of nano particles with ratio of (0.01 Al + 0.03 ZrO2) to the Nano composite material (PMMA: 0.03 fly ash) (fig. 14) and to (PMMA:X% fly dust) with (0.01, 0.02 and 0.03 fly dust) (Fig. 15) and the addition of the mixture of nano particles with ratio of (0.01 fly ash + 0.03 fly dust) to the nano composite material (PMMA: 0.03 nZrO2) (fig. 16) leads to decrease in the

compressive strength of the PMMA hybrid nano composites after exposure to UV radiation and moisture as comparing with net PMMA. The reasons behind such a behavior are related to the same reasons, which mentioned in the previous item for figures 12 and 13.

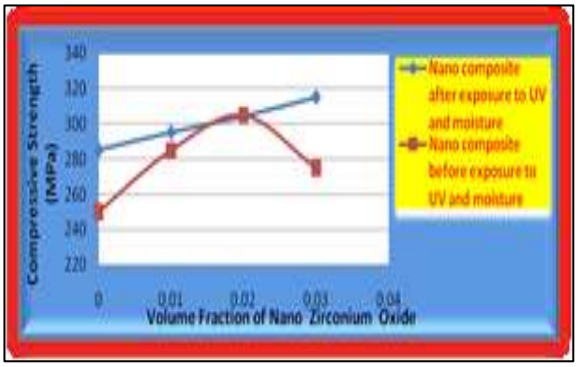


Figure 13: Compressive strength of PMMA nano composite materials either before or after exposure to accelerated weathering as a function of volume fraction of zirconium oxide nano particles in PMMA matrix

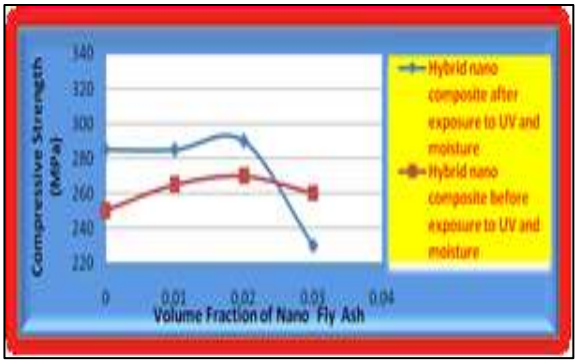


Figure 14: Compressive strength of PMMA hybrid nano composites as a function of volume fraction of fly ash nanoparticles in PMMA composites

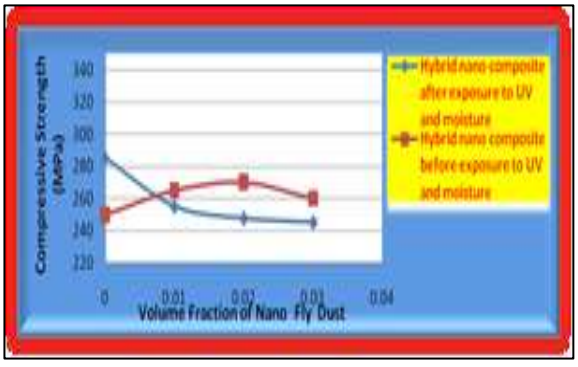


Figure 15: Compressive strength of PMMA hybrid nano composites as a function of volume fraction of fly dust nano particles in PMMA composites

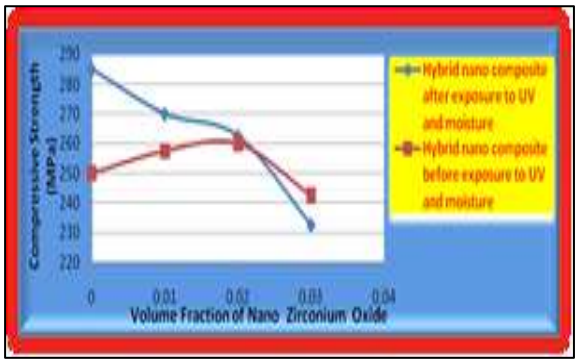


Figure 16: Compressive strength of PMMA hybrid nano composites as a function of volume fraction of zirconium oxide nano particles in PMMA composites

6. Conclusions

In the present work, attempts are made to development PMMA polymer, which is used in the denture base and in dental prostheses applications. So the nano composites and hybrid nano composites with desirable properties were attended, by adding three types of nano powders (fly ash, fly dust and ZrO_2) at the same ratio to it, as well as effect of UV radiation and moisture also studied, and it was concluded the following:

• The compressive strength and compressive elastic modulus increased with the addition low volume fractions of the nanoparticles (fly ash, fly dust, aluminum and zirconium oxide) and decreased with the addition high volume fractions to larger than 0.02 of the nanoparticles to the PMMA nano composites and PMMA hybrid nano composites.

• The addition of ZrO2 nanoparticles has a noticeable effect on the compressive strength, under the effect of UV radiation and moisture of the nano composite and hybrid nano composite prosthetic denture base specimens more than the fly ash and fly dust nanoparticles.

• The maximum value for the compressive strength and compressive elastic modulus was obtained in the nano composite material (PMMA - 0.02 nZrO2).

• The maximum value for the compressive strength under the effect of UV radiation and moisture was obtained in the nano composite material (PMMA - 0.03 nZrO2). Based on the foregoing, and in order to stabilizing and increasing in the compressive strength of PMMA nano composites and PMMA hybrid nano composites under the effect of UV radiation and humidity, it should be selected the (PMMA- 0.01 and 0.02 fly ash), (PMMA- 0.01 fly dust) and (PMMA - 0.01, 0.02 and 0.03 nZrO2) as a nano composites and ((PMMA: 0.02 fly ash) - (0.01 Al+0.03 ZrO2)) as a hybrid nano composite

because with these constituents make the PMMA nano composites and PMMA hybrid nano composite stable and increase its compressive strength after exposure to UV radiation and moisture as comparing with pure PMMA by preventing the degradation process of the PMMA nano composites and PMMA hybrid nano composites.

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