COMPARATIVE STUDY THE FLEXURAL PROPERTIES AND IMPACT STRENGTH FOR PMMA REINFORCED BY PARTICLES AND FIBERS FOR PROSTHETIC COMPLETE DENTURE BASE

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ABSTRACT :-

In the present search, attempts are made to develop the properties of PMMA resin that used for upper and lower prosthesis complete denture, by addition two different types of particles, which included: nano-hydroxyapatite (nHA) particles and micro-zirconia (ZrO_2) particles that added with different volume fractions of (1%, 2% and 3%) to poly methyl methacrylate (PMMA) cold cured resin as new fluid resin matrix. Also woven glass fiber kind (E-glass) and woven Kevlar fiber kind (49), it were added with a fixed volume fraction of (5%) to PMMA composites. In this work the composite prosthetic dentures specimens preparation was done by using (Hand Lay-Up) method as six groups which includes: the first group consists of PMMA resin reinforced by nHA particles, the second group consists of PMMA resin reinforced by ZrO_2 particles, the third group consists of (PMMA-nHA) and glass fiber layer as laminated composite , the fourth group consists of (PMMA-ZrO_2) and glass fiber layer, the fifth group consists of (PMMA-nHA) and Kevlar fiber layer and the sixth group consists of (PMMA-ZrO_2) and Kevlar fiber layer.

The experimental part of this study included performing many mechanical tests which, includes (Flexural, Maximum Shear Stress and Impact) tests. The result of this study showed the values of most properties increased with increasing of the volume fraction of (nHA and ZrO_2) particles in polymer composite materials. While, the impact strength decreased. The ZrO_2 particle and woven mat Kevlar fiber had shown greater values for the most composite materials properties for all groups' specimens except the flexural strain.

Also the results showed the maximum values for hybrid laminated composite properties (flexural modulus and impact strength) were obtained in hybrid laminated composite materials for sixth group specimens. While, the maximum value for properties (flexural strength and max shear stress) were obtained in hybrid laminated composite materials for fourth group specimens.

Key Words: PMMA, Nano-HA Particles, ZrO_2 Particles, Glass Fibers, Kevlar Fibers, Hybrid Composite, Flexural Modulus, Flexural Strength, Max Shear Stress, Impact Strength.

دراسة مقارنة حول خصائص الانحناء ومقاومة الصدمة للبولي مثيل ميثا اكريليت المقوى بالدقائق والألياف لمادة قاعدة طقم أسنان أصطناعي كامل

الخلاصة :

تم في هذا البحث أجراء عدة محاولات لغرض تطوير مواصفات راتنج البولي مثيل ميثا اكريليت المستخدم كطقم أسنان أصطناعي كامل علوي أو سفلي. وقد حضرت هذه المواد المتراكبة من البولي مثيل ميثا اكريليت على شكل راتنج سائل جديد معالج ذاتياً كمادة أساس وقد تم تقويتها بنوعين مختلفين من الدقائق تضمنت دقائق الهيدروكسي اباتايت النانوية ودقائق الزركونيا الميكروية اضيفت بكسور حجمية مختلفة هي (1%, 2% and 3%)، ونوعين مختلفين من الالياف الحصيرية الثنائية الاتجاه هي الياف الزجاج نوع (E-GLASS) والياف الكفلر نوع (49) وقد اضيفت الى المواد المتراكبة بكسر حجمي ثابت هو (5%). في هذا البحث حضرت عينات من أطقام الاسنان الأصطناعية المتراكبة بطريقة (الصب اليدوي) على شكل ستة مجاميع هي: المجموعة الاولى تضم راتنج البولي مثيل ميثا اكريليت ودقائق الهيدروكسي اباتايت النانوية، المجموعة الثانية تضم راتنج البولي مثيل ميثا اكريليت ودقائق الزركونيا، المجموعة الثالثة تضم راتنج البولي مثيل ميثا اكريليت والياف الزجاج مع دقائق الهيدروكسي اباتايت النانوية، المجموعة الرابعة تضم راتنج البولي مثيل ميثا اكريليت والياف الزجاج مع دقائق الزركونيا، المجموعة الخامسة تضم راتنج البولي مثيل ميثا اكريليت والياف الكفلر مع دقائق الهيدروكسي اباتايت النانوية، المجموعة السادسة تضم راتنج البولي مثيل ميثا اكريليت والياف الكفلر مع دقائق الزركونيا.

يتضمن الجزء العملي من هذا البحث اجراء العديد من الاختبارات الميكانيكية والتي شملت فحوصات (الانحناء، اجهاد القص الأعظم والصدمة). وقد بينت النتائج بان قيم اغلب الخواص ازدادت مع زيادة الكسر الحجمي لدقائق الهيدروكسي أباتايت النانوية ودقائق الزركونيا، بينما قلت مقاومة الصدمة. وان دقائق الزركونيا والياف الكفلر الحصيرية تعطي قيم أعلى لاغلب خواص المواد المتراكبة ولكافة مجاميع العينات ماعدا الانفعال الأنحنائي.

كذلك أظهرت النتائج بان اعلى قيم لخصائص (معامل الانحناء ومقاومة الصدمة) وجدت في المواد المتراكبة الطبائقية الهجينية لعينات المجموعة السادسة. بينما اعلى قيم لخصائص (مقاومة الانحناء واجهاد القص الأقصى) وجدت في المواد المتراكبة الطبائقية الهجينية لعينات المجموعة الرابعة.

1- INTRODUCTION

The composite materials have many important mechanical properties which make them suitable for many industrial uses. This is approach the scientist to study the effected of different type of reinforced phase on the mechanical properties for the composite material, such as (flexural strength, flexural modulus, impact strength, etc) some researches which are accomplished in this field it's:-

Vallittu et. al., (1995), investigated the properties of the unidirectional denture glass fiber-poly methyl methacrylate (GF-PMMA) denture composite and compared with the continuous glass fiber or metal wire. The impact strength, tensile strength and modulus of elasticity for the test specimens were measured. The results showed that both types of reinforcement increased the impact strength if compared with unreinforced PMMA. However, there was no clear difference between mean impact strength values of specimens reinforced with metal or glass fiber.

Teoman et. al., (1999), measured the effect of five types of fiber strengtheners on the fracture resistance of denture base acrylic resin material. Impact strength, transverse strength, deflection, and elasticity modulus values of a heat- polymerized denture base resin reinforced with glass, carbon, thin Kevlar, thick Kevlar, and polyethylene fibers in woven form were studied. The results were highest impact test values produced by polyethylene fibers reinforced group, and the lowest values were obtained from specimens non reinforced fibers. There were significant differences test group specimens, but no significant in transverse strength between specimens strengthened with polyethylene fibers and non reinforced fibers.

Kanie et. al., (2000), investigation the reinforcing effect of woven glass fibers on deflection, flexural strength, flexural modulus and impact strength of three acrylic resin denture base silanized or unsilanzied woven glass fibers was used. Specimens were made by heating the denture cure resin dough containing glass fiber. The flexural strength and deflection in specimens reinforced with silanized glass fiber of (1 mm) thickness were significantly higher than those of unreinforced specimens. Furthermore, the impact strength in specimens reinforced with silanized glass fiber of (2 mm) thickness was significantly higher than that of unreinforced specimens.

Tacir et. al., (2006), studied the effect of glass fiber reinforcement on fracture resistance and flexural strength of acrylic denture base resin, this study was indicated the possibility for improving the flexural strength of heat polymerized PMMA after reinforcement with glass fiber, and may be possible to apply distal tension on partial and complete denture bases.

Ali S., (2006), investigate the problem of radio- opacity by incorporating titanium powder as a radio- opaque fillers and study the effect of these additive fillers (Ti powder and ZrO_2 powder) on some properties of hot cure PMMA denture base material. It was found the addition of Ti powder slightly increased transverse strength, but slightly decreases impact strength. The additions of ZrO2 powder highly decrease both impact strength and transverse strength.

Chow W. S. et. al., (2008), prepared PMMA/HA composites by using heat- processing polymers powder and liquid method. The flexural modulus of PMMA was increased by the addition of HA this is attributed to the reinforced effect of HA. However, the flexural strength and flexural strain was decreased in the presence of HA.

Azlan et. al., (2008), attempted to investigate the effects of hydroxyapatite (HA) on the flexural properties, fracture toughness, and glass transition temperature (Tg) of PMMA powder was mixed with monomer of methyl methacrylate (MMA). The PMMA/HA composites were prepared by using heat- cure PMMA resin, the polymerization of PMMA/HA were carried using water bath. It was found that the young modulus, fracture toughness and Tg properties of PMMA were influenced by the HA addition.

Komar et. al., (2009), assessed the flexural strength values of denture base polymers reinforced with glass fibers after performed artificial ageing. The specimens after polymerization immersed in water of temperature (37 ºC) for 28 days. The results flexural strength of the polymer specimens was (91.76-122.75 MPa), while flexural strength values of specimens reinforced with glass fibers (103.10-163.88 MPa).

Ihab, (2011), evaluated the effect of addition of modified nano-zirconium oxide (ZrO_2) particles on some properties of heat cure acrylic denture base material. Nano-ZrO_2 was coated with a layer of tri methoxy sily prople methacrylate (TMSPM) before dispersed in monomer (MMA) in different percentages (2%, 3%, 5% and 7%) by weight. It was found that the addition of modified nano-ZrO_2 highly increase the radio-opacity of heat cure PMMA and this increase were proportional to concentration of nano-ZrO_2. The maximum increase in impact and transverse strength was observed in nano-composite containing (5wt %) of nano-ZrO_2 and then strength decreases with further increase.

Chow W. S. et. al, (2012), surface treatment of HA using zirconate agent (ZCA) carried out to improve the interfacial bonding between the PMMA matrix and HA filler. The mechanical properties of PMMA/HA composites was investigated using flexural tests. It was found that (PMMA-5% HA-2% ZCA) composites exhibited higher flexural strength compared to that of untreated PMMA/HA composites. This is attributed to the enhancement of interaction between PMMA and HA.

The aim of this research is studying the effect of selected volume fractions of (nHA and ZrO_2) particles and (woven glass, woven Kevlar) fibers on the flexural and impact properties of the composite and hybrid composite prosthetic complete denture.

2- MATERIALS AND METHODS :-

2-1 Materials Used

In this research the composite prosthetic dentures specimens consist of polymer matrix and reinforced materials (relatively high stiffness and high wear resistance).

Matrix Material included PMMA cold curing that used in this research as new pour (fluid) resin matrix, type (Castavaria) made from (Vertex – Dental Company), to preparation specimen as hybrid and non hybrid laminated composites of the denture prosthetic. Vertex™ Castavaria is a multifunctional self polymerizing acrylic which is perfectly useable as a pouring, relining, rebasing and as a repair acrylic.

This type of materials distinguishes by many properties compared with other type of PMMA polymer such as: softer feel, low molecular weight, color stable in the long run, minimized shrinkage, stable polymerization cycle with a perfect end result, the acrylic is long pourable and modelable for a long period of time. But have low strength, low hardness and more difficult using during fabrication (William and O' Brain, 2002).

Two types of particles were used in this study as reinforces materials with volume fraction of (1%, 2% and 3%) it was added to the polymer (acrylic powder) including: the zirconium oxide (ZrO_2) is supplied as partially stabilized particles, which made from (ZIRCONIA SALES-GUI 185 SS-U.K Company). The result of particle size distribution of (ZrO_2) particles is obtained by using AFM which shows the average diameter was (112.31 nm). The result of particle size distribution of (ZrO_2) particles is shown in the Figure (1). Table (1) shows the mechanical and physical properties of this particle.

The HA particles most common widely used in medical application and supplied as a nano-particles and represented as chemical formula $Ca_5\ (PO_4)_3\ OH$ which made from (Merck, Darmstadt, Germany Company). The result of particle size distribution of (HA) particles is obtained by using AFM which shows the average value diameter was (69.97 nm). The result of particle size distribution of (HA) particles is shown in the Figure (2). Table (2) shows the mechanical and physical properties of this particle.

Two types of fibers were used in this study including: E-glass fiber used in this research made from (Mowding LTD-U.K Company), as form a woven mat with fibers angle direction ($0^{o}/90^{o}$) and volume fraction (5%). The Kevlar 49 fiber used in this research made from (E.I.Dupont de. Nomours Company), as form a woven mat with fibers angle direction ($0^{o}/90^{o}$) and volume fraction (5%). This type of fiber has a yellow color. This color changes according to methods of its yarn.

2-2 Preparation Methods of Test Specimens

2-2-1 Proportioning and Mixing of Acrylic

The Vertex™ Castavaria was used to prepare the specimens of the PMMA composite materials. The standard proportion in mixing ratio is usually for cold cure acrylic resin of (17 g) polymer powder (PMMA) and (10 ml) monomer liquid (MMA) (1.7 g / 1 ml) by volume or (1.7g / 0.95g) by weight according to the manufacturer's instructions of manufacturer company.

The importance of this ratio was related to control the workability of the mixture, dimensional changes on setting and considered one of the variables influence the cytotoxicity of acrylic resin. When mixing powder and liquid many changes will take place due to solution of polymer in the monomer (Jorge. J. H., 2003).

The Vertex Castavaria is mouldable for a long period of time, where the mixture was mixed of liquid (MMA) in the clean and dry container (glass beaker), follow after that by slow addition of dry powder (PMMA) to liquid (MMA), the mixture was stirred at room temperature continuously by using mechanical mixing (brabender plastograph mixer) at speed (20 r.p.m.) until reached to the dough stage and poured with thin straight line in the center of opening mould with maximum time about (4.5 min) according to manufacture company. During mixture pouring in the glass mould, the mould must be rocked very gently and vibrated from side to side to remove any gas bubbles from the specimens, and reminder of the mixture was poured into mould hole until the glass mould filling. This mixture was covered in closed container and left to stand on the bench top at room temperature (23 ± 2) ºC for (8-13) min from beginning of mixing process as working time to increase the viscosity of mixture and surface of the pouring has become hard and matt.

2-2-2 Curing Cycle Employed

According to the manufacture's instruction polymerization curing the closed mould was placed in the pressure vessels (autoclave). Therefore all specimens were then placed inside autoclave at (55 ºC) and pressure equal to (2.5 bar) and let for (30 min) according to manufacturer's instructions of manufacturer company, because of the specimens' complete polymerization under this condition. The advantage of this technique is polymerization may be accomplished in short time, post cured of specimens and give minimum level of residual monomer.

2-2-3 Cooling Process

After polymerization completion follow by curing process, the mould was allowed to stand outside of the autoclave and put on the bench for cooling time about (30 min) at room temperature to complete the cooling and complete hardening of specimen. After cooling, the specimens were de-mould to remove from the mould and cleaning. Figures (3 a, b and c) show the some casting specimens after de-mould process.

3- EXPERIMENTAL TESTS

Some mechanical tests were performed in this study to evaluate the flexural and impact properties of the PMMA composite materials and hybrid laminated composite materials of the denture prosthetic which includes the flexural test is performed according to (ASTM D790) and (ADA Specification No.12, 1999) at room temperature (23 ± 2) ºC. All data in this test were measured from three point bending test machine by using tensile machine at across head speed (strain rate) of (5 mm/min) and load was applied equal to (5KN) until break the specimen occur (Annual Book of ANSI/ADA Standard, 1999 and Annual Book of ISO Standard, 2006).

The impact test is performed according to (ISO-180) and (ADA Specification No.12, 1999) at room temperature (23 ± 2) ºC by using Izod Impact test machine type is (XJU series pendulum Izod/Charpy impact testing machine). For Izod test: the specimen 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). Unnotched Izod impact is a single point test that measures a material resistance to impact from a swinging pendulum.

Also the shear test is performed according to (ASTM D2344) and (ADA Specification No.12, 1999) at room temperature (23 $\pm$ 2) ºC. All data measured from three points bending test

machine by using (Hydraulic press) type (Leybold Harris No. 36110) with used short beam and gradually load applied.

All these tests carried out in air after immersion the specimens in distilled water at (37 $\pm$ 1) ºC for (48 hr), five times were tested for each specimen and take the averaged for the final result (Annual Book of ASTM Standard, 2003 and Annual Book of ASTM Standard, 1997)

4- RESULTS AND DISCUSSIONS :-

4-1 Results and Discussions of the Flexural Test for Modified Composites

The flexural modulus values results obtained from flexural test that carried out on PMMA composite are discussed in Figures (4), (5) and (6) respectively.

From Figure (4) can be noticed that the values of flexural modulus increased with increasing of the volume fraction of both types of particles in the PMMA composite materials. This is due to the strengthening mechanism and nature of bonding, reinforced particles and interface between reinforcing materials and PMMA matrix. Also this increase may be due to the fact of high flexural modulus value of the (nHA and ZrO_2) particles as compared with PMMA matrix (S. M. Elie., 2007).

Also can be noticed in this Figure that the values of flexural modulus for second group (ZrO_2-PMMA) composite specimens are higher than values their counter parts of the first group (nHA-PMMA) composite specimens, and increased with a higher rate comparing with the behavior of first group specimens. This is due to the improvement of the mechanical properties that associated with the addition of ZrO_2 particles to PMMA that related to the nature of ZrO_2 particles which have high flexural modulus comparing with HA particles as seen from Tables (1) and (2). Thus, the flexural modulus values increased from (2.05 GPa) for PMMA (as referenced) to (8.63 GPa) for (PMMA-3% ZrO_2) composite.

From the Figures (5) and (6) can be seen that when adding of woven mat of glass fiber or Kevlar fiber in PMMA composite, the flexural modulus increased with increasing (nHA and ZrO_2) contact in composites with symmetrically manner approximately. This could be attributed to the fact that glass fiber and Kevlar fiber are characterized by their higher flexural modulus than PMMA matrix. Therefore, that leads to improving the flexural modulus of the hybrid laminated composite specimens.

And can be noticed in Figures (5) and (6) that the values of flexural modulus increased with increasing volume fraction of (nHA and ZrO_2) particles for all hybrids laminated composite specimens, this is due to the contribution of either (nHA or ZrO_2) particles, glass fiber and Kevlar fiber to be carried out of the load applied on the hybrid laminated composite specimens that suitable their mechanical properties, nature and geometry as well as their volume fraction. Therefore, the using of (HA and ZrO_2) particles which have flexural modulus more than other reinforced material such as (glass fiber and Kevlar fiber) and PMMA matrix materials which have low flexural modulus. As well as the regular and randomly distribution of (nHA and ZrO_2) particles inside the PMMA resin which that evidenced by using SEM test, and ease penetration the matrix material through the particles and woven mat fibers to formation good adhesion at interfaces between the matrix and reinforcing material, all these reasons lead to increase of flexural modulus with every increasing in volume fraction of (nHA and ZrO_2) particles (Henning K. et. al., 2001).

The SEM images of the surface morphology of the (PMMA-3% ZrO_2) composite, (PMMA-3% ZrO_2-5% Glass fiber) composite and (PMMA-3% ZrO_2-5% Kevlar fiber) composite are shown in Figures (7 a, c and e) respectively, but the surface morphology of the (PMMA-3% nHA) composite, (PMMA-3% nHA-5% Glass fiber) composite and (PMMA-3% nHA-5% Kevlar fiber) composite are shown in Figures (7 b, d and f) respectively. From these Figures it is found there is a difference in the surface morphology of the (nHA-PMMA) composites when compared with (PMMA- ZrO_2) composites. The dark regions are PMMA phase while the bright regions are reinforcement phase (HA or ZrO_2) particles.

Moreover, these Figures show that the values of flexural modulus for the hybrid laminated composite specimens reinforced by woven mat Kevlar fiber are higher than values compared with the values of flexural modulus for the hybrid laminated composite specimens reinforced by woven mat glass fiber with every increase in the volume fraction of (nHA or ZrO_2) particles. This is due to the characteristics that distinguished the Kevlar fiber compared to glass fiber in which the former has high flexural modulus than the latter. This is due to the improvement of the mechanical properties that are associated with the addition of ZrO_2 particles to hybrid laminated composite specimens. Thus, the higher values of flexural modulus reach to (11.18 GPa) for hybrid laminated composites (PMMA-5% Kevlar fiber-3% ZrO_2). Finally, the percentage improvement was (81.66 %).

While the flexural strain values results which obtained from flexural test that carried out on PMMA composite materials and hybrid laminated composite materials for all groups specimen that prepared in this research are shown in Figures (8), (9) and (10) respectively.

From the Figure (8) can be noticed that the values of flexural strain decreased with increasing of the volume fraction of both types of particles for both groups of PMMA composite materials symmetrically. This is because of any increasing in the particle number, it will be act as localized stress concentration regions. Therefore, the flexural strain will be decreased. Also the decreasing in the flexural strain depends up on the interfaces bonding between the PMMA matrix and reinforcing material (nHA and ZrO_2) particles therefore the decreasing in the flexural strain may be attributed to the formation strong structure of the PMMA composite material for specimen of this groups (Screekanth M. S. et. al., 2009 and Sawalha S. et. al., 2007).

It can also be noticed in this Figure that the values of flexural strain for second group (ZrO_2-PMMA) composite specimens are lower than the values of flexural strain for first group (nHA-PMMA) composite specimens. This is due to the higher mechanical strength (flexural strength) of ZrO_2 particles comparing with HA particles and then decreases the flexural strain. Thus, the flexural strain value decreased from (0.0413 %) for PMMA (as referenced) to (0.0136 %) for (PMMA-3% ZrO_2) composite.

From the Figures (9) and (10) can be seen that when adding of woven mat of glass fiber or Kevlar fiber in PMMA composite, the flexural strain increases. This could be attributed to the fact that glass fiber and Kevlar fiber are characterized by their higher flexural strain than PMMA matrix. Therefore, that leads to improving the flexural strain of the hybrid laminated composite specimens.

And can be noticed in Figures (9) and (10) that the values of flexural strain increased with increasing of the volume fraction of (nHA and ZrO_2) particles for all hybrids laminated composite specimens. This is due to the regular and randomly distribution of (nHA and ZrO_2) particles inside the PMMA matrix and good ability of PMMA resin to penetration of these particles and woven mat (glass and Kevlar) fibers and spread on the (nHA and ZrO_2) particles also ease penetrate of these particles inside the interstitial voids and space that was found in woven mat fibers without make any defects inside them, and increasing the wettability of reinforced materials this is lead to create perfect interact between the matrix and reinforcing

materials and increasing the bond strength between them which that evidenced by using SEM test,, then they diminish these discontinuous regions which act as stress raiser and diminish the stress concentration region therefore , all these reasons lead to increase the values of flexural strain for all hybrid laminated composite materials groups with every increasing in the volume fraction of (nHA and ZrO_2) particles (C. A. Harper, 2006).

Moreover, these Figures show that the values of flexural strain for the hybrid laminated composite specimens reinforced by woven mat glass fiber are higher than values compared with the values of flexural strain for the hybrid laminated composite specimens reinforced by woven mat Kevlar fiber with every increase in the volume fraction of (nHA or ZrO_2) particles. Also the specimens which have glass fibers appear high flexural strain under the load applied before fracture occur. This is due to the nature and the characteristics that distinguished the glass fibers compared to Kevlar fiber in which the former has high flexural strength and flexural strain than the latter, in addition to the Kevlar fiber have anisotropic properties and weakness toward flexural stress and Max. shear stress.

In addition to, Figures (9) and (10) show that the values of flexural strain increased when adding HA particles of the hybrid laminated specimens for the third and fifth groups more than the values of Flexural Strain when adding ZrO_2 particles to the hybrid laminated specimens for the fourth and sixth groups. This is due to the high mechanical Strength (flexural strength) that is associated with the addition of ZrO_2 particles to hybrid laminated composite specimens comparing with HA particles and then decrease the flexural strain. Thus the higher values of flexural strain reach to (0.1744 %) for hybrid laminated composite (PMMA-5% Glass fiber-3% nHA).

4-2 Results and Discussions of the Flexural Strength and Maximum Shear Strength Test for Modified Composites .

The failure of composite material occurs as an effect one of these stresses depending upon the type of reinforcing materials and the bond strength between the matrix and reinforced materials. The flexural strength, max. shear strength values results obtained from flexural test that carried out on PMMA composite are discussed in Figures (11), (12), (13), (14), (15) and (16) respectively

From Figure (11) and (12) can be noticed that the values of flexural strength and max. shear strength increased with increasing of the volume fraction of both types of particles in the PMMA composite materials. This is due to the ability of these particles to hinder the crack propagation inside PMMA matrix according to strengthening mechanism additionally to the strong bonding between the PMMA matrix and these particles. Furthermore, this increasing may be due to the fact the flexural strength and max. shear strength of (HA and ZrO_2) particles are much higher than PMMA matrix (B. R. Varadharajan, et. al., 2005).

It can also be noticed in these Figures that the values of flexural strength and max. shear strength for second group (ZrO_2-PMMA) composite specimens are higher than values their counter parts of the first group (nHA-PMMA) composite specimens, and increased with a higher rate comparing with the behavior of first group specimens. This is due to the improvement of the mechanical properties that are associated with the addition of ZrO_2 particles to the PMMA that related to the nature of ZrO_2 particles which have high flexural strength and max. shear strength comparing with HA particles as seen from Tables (1) and (2). Thus, the flexural strength and

max. shear strength values increased from (80.792 and 4.59 MPa) for PMMA (as referenced) to (143.498 and 8.153 MPa) respectively for (PMMA-3% ZrO_2) composite.

From the Figures (13), (14), (15) and (16) can be seen that when adding of woven mat of glass fiber or Kevlar fiber in PMMA composite, the flexural strength and the max. shear strength increasing to the double values of PMMA matrix with increasing (nHA and ZrO_2) contact in composites with symmetrically manner approximately. This could be attributed to the fact that glass fibers and Kevlar fiber are characterized by their higher flexural strength and max. shear strength than PMMA matrix. Therefore, that leads to improving the flexural modulus of the hybrid laminated composite specimens.

And can be noticed in these Figures that the values of flexural strength and max. shear strength increased with increasing volume fraction of (nHA and ZrO_2) particles for all hybrids laminated composite specimens, this is due to the higher bond strength that may occur between the fibers and matrix. In addition to the reasons that explanation about Figures (4) and (5) for the flexural modulus, all these reasons lead to improving the flexural strength and max. shear strength of the hybrid laminated composite specimens for all groups with increasing in the volume fraction of (nHA and ZrO_2) particles (P. K. Mallick, 2008).

Furthermore, it can be noticed in these Figures that the values of flexural strength and max. shear strength respectively, for the hybrid laminated composite specimens reinforced by woven mat glass fiber are higher than values compared with the values of flexural strength and max. shear strength for the hybrid laminated composite specimens reinforced by woven mat Kevlar fiber with every increase in the volume fraction of (nHA or ZrO_2) particles. This is due to the nature and the characteristics that distinguished the glass fibers compared to Kevlar fiber in which the former has high flexural strength as well as a high max. shear strength than the latter, in addition to the Kevlar fiber have anisotropic properties and weakness toward flexural strength and max. shear strength, whereas the glass fibers have isotropic properties and be strong toward the flexural strength and shear strength. In addition to, the improvement of the mechanical properties that are associated with the addition of ZrO_2 particles to hybrid laminated composite.

Thus, the higher values of flexural strength and max. shear strength reach to the (239.632 and 13.615 MPa) for hybrid laminated composite (PMMA-5% Glass fiber-3% ZrO_2). Finally, the percentage improvement were (66.28 %) for the flexural strength and (66.29 %) for the Max. shear stress.

4-3 Results and Discussions of the Impact Test for Modified Composites

The impact strength values results obtained from impact test that carried out on PMMA composite and hybrid laminated composite materials for all groups specimen that prepared in this research are shown in Figures (17), (18) and (19) respectively.

From the Figure (17) can be noticed that the values of impact strength decreased with increasing of the volume fraction of both types of particles with symmetrically manner approximately for both groups of PMMA composite materials. This is because of any increasing in these particles numbers, it will be act as points for localized stress concentration regions from which the failure will begin. Furthermore, this decreasing may be due to the fact of these particles brittleness and weakness in the ability of resistance to impact load comparing with PMMA matrix. Therefore, the impact strength will be decreased.

It can also be noticed in this Figure that the values of impact strength for first group (nHA-PMMA) composite specimens are lower than the values of impact strength for second group (ZrO_2-PMMA) composite specimens. This is due to the higher hardness of ZrO_2 particles comparing with HA particles and then decreased the impact strength.

Thus, the impact strength value decreased from (9.56 KJ/m^2) for PMMA (as referenced) to (6.43 KJ/m^2) for (PMMA-3% nHA) composite.

From the Figures (18) and (19) can be seen that when adding of woven mat of glass fiber or Kevlar fiber in PMMA composite, the impact strength increasing as compared with Figure (16) for PMMA matrix composite. This could be attributed to the fact that glass fiber and Kevlar fibers are characterized by their higher impact strength than PMMA matrix, therefore that lead to improving the impact strength of the hybrid laminated composite specimens.

It can also be noticed in these Figures that the values of impact strength decreased with increasing of the volume fraction of (nHA and ZrO_2) particles for all hybrids laminated composite specimens. This is due to the brittleness of these particles and weakness in the ability to resistance of the impact load compared with the other components of the composite materials which include the PMMA, glass fibers, and Kevlar fibers, also these particles act as points a localized stress concentration to allow failure begin.

Furthermore, it can be noticed in Figures (18) and (19) that the values of impact strength for the hybrid laminated composite reinforced by woven mat Kevlar fiber are higher than values compared with the values of impact strength for the hybrid laminated composite reinforced by woven mat glass fiber with every increase in the volume fraction of (nHA or ZrO_2) particles. This is due to the nature and the characteristics that distinguish the Kevlar fibers compared to glass fiber in which the former has high toughness and high impact strength compared than the latter, besides has brittleness and easily broken into small pieces. Thus, the impact strength decreased to lower value (21.05 KJ/m^2) for hybrid laminated composite (PMMA-5% Glass fibers-3% ZrO_2). Finally, the percentage improvement was (84.66 %).

5- CONCLUSIONS :-

From the presented experimental results of this research, the following can be the conclusions:

1- All mechanical properties of PMMA composites (PMMA-nHA), (PMMA-ZrO_2) and hybrid laminated composites specimens, increased with increasing of the volume fractions of (nHA and ZrO_2) particles, except the (impact strength) values was decreased with increasing of the volume fraction of (nHA and ZrO_2) particles.

2- The addition of ZrO_2 particles has a noticeable effect on the most properties of composite prosthetic denture base for all groups' specimens more than the HA particles. While, the (flexural strain) has higher values when added HA particles.

3- The maximum values for properties (flexural modulus and impact strength) were obtained in hybrid laminated composite materials (PMMA-K.F-ZrO_2).

4- The maximum values for properties (flexural strength and max shear stress) were obtained in hybrid laminated composite materials (PMMA-G.F-ZrO_2).

5- The maximum values for flexural strain property were obtained in hybrid laminated composite materials (PMMA-G.F-nHA).

(Table 1): Some Mechanical and Physical Properties of ZrO_2 Particles that used in this Study according to (ZIRCONIA SALES-GUI 185 SS-U.K Company).

Type of Particle	Tensile Strength (MPa)	Young's Modulus (GPa)	Possion's Ratio	Compression Strength (MPa)	Flexural Strength (MPa)	Density (gm/cm^3)
Zirconium Oxide	800-1500	205-210	0.23-0.31	2000	900-1200	5.7-6.1

(Table 2): Some Mechanical and Physical Properties of H.A Particles that used in this Study according (Merck, Darmstadt, Germany Company).

Type of Particle	Tensile Strength (MPa)	Young's Modulus (GPa)	Possion's Ratio	Compression Strength (MPa)	Flexural Strength (MPa)	Density (gm/cm^3)
Hydroxyapatite	40-100	70-120	0.28	100-900	20-80	3.08-3.18

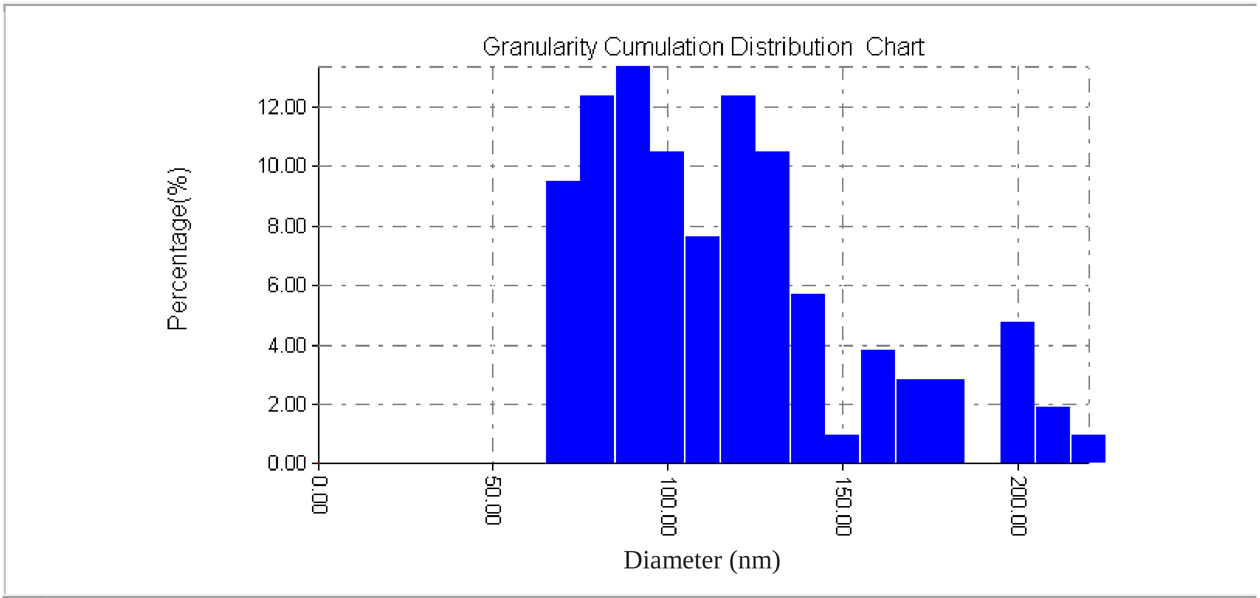
Scanning Probe Microscope

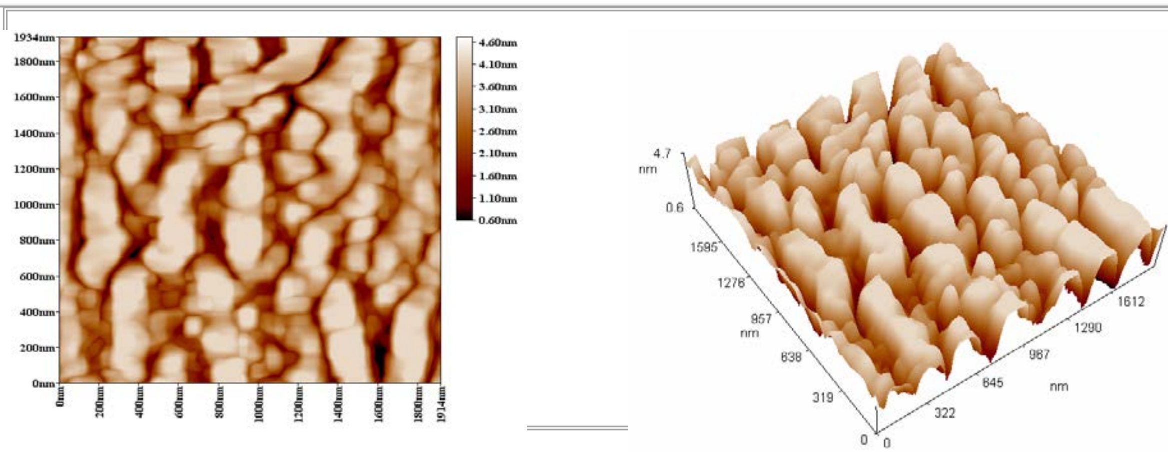
Granularity Cumulation Distribution Report

Sample: ZRO_2	**Code:** Sample Code
Line No.: lineno	**Grain No.:** 105
Instrument: CSPM	**Date:** 2013-02-09

Avg. Diameter: 112.31 nm

Diameter(nm)<	Volume (%)	Cumulation(%)	Diameter(nm)<	Volume (%)	Cumulation(%)	Diameter(nm)<	Volume (%)	Cumulation(%)
70.00	9.52	9.52	120.00	12.38	65.71	170.00	2.86	89.52
80.00	12.38	21.90	130.00	10.48	76.19	180.00	2.86	92.38
90.00	13.33	35.24	140.00	5.71	81.90	200.00	4.76	97.14
100.00	10.48	45.71	150.00	0.95	82.86	210.00	1.90	99.05
110.00	7.62	53.33	160.00	3.81	86.67	220.00	0.95	100.00





(Fig. 1): AFM Test of ZrO_2 Particles.

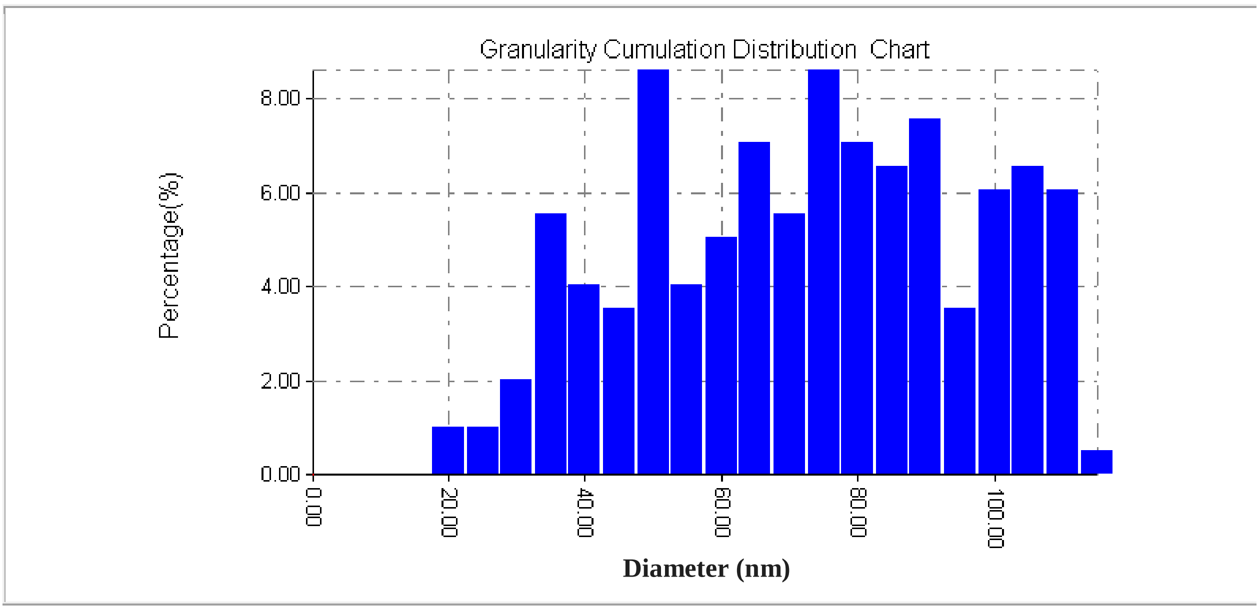
Scanning Probe Microscope

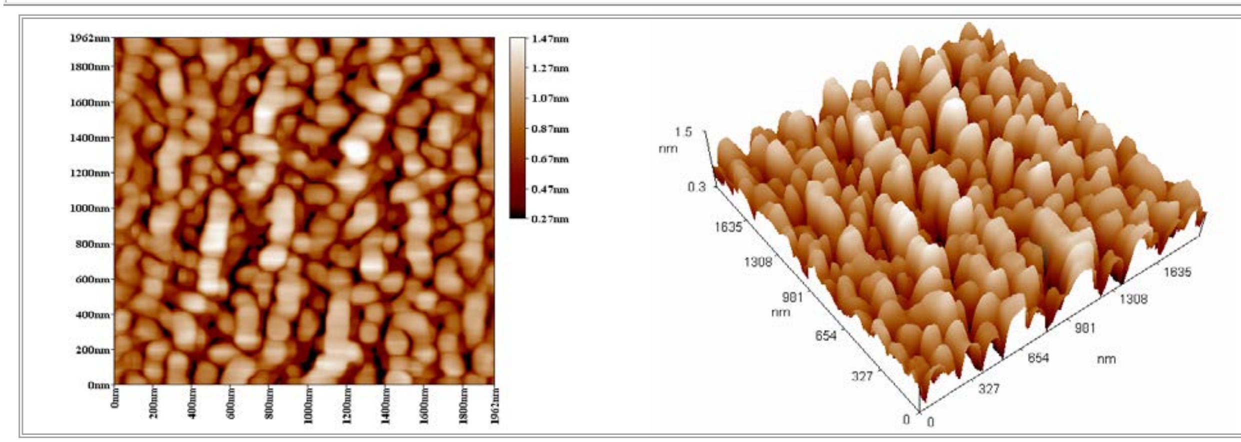
Granularity Cumulation Distribution Report

Sample:HA	**Code:**Sample Code
Line No.:lineno	**Grain No.:**198
Instrument:CSPM	**Date:**2013-02-09

Avg. Diameter:69.97 nm

Diameter(nm)<	Volume(%)	Cumulation(%)	Diameter(nm)<	Volume(%)	Cumulation(%)	Dia
20.00	1.01	1.01	55.00	4.04	29.80	
25.00	1.01	2.02	60.00	5.05	34.85	
30.00	2.02	4.04	65.00	7.07	41.92	
35.00	5.56	9.60	70.00	5.56	47.47	
40.00	4.04	13.64	75.00	8.59	56.06	
45.00	3.54	17.17	80.00	7.07	63.13	
50.00	8.59	25.76	85.00	6.57	69.70	

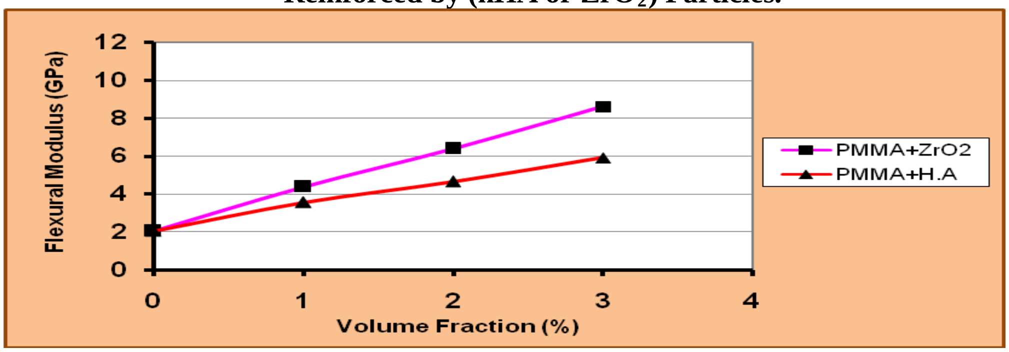




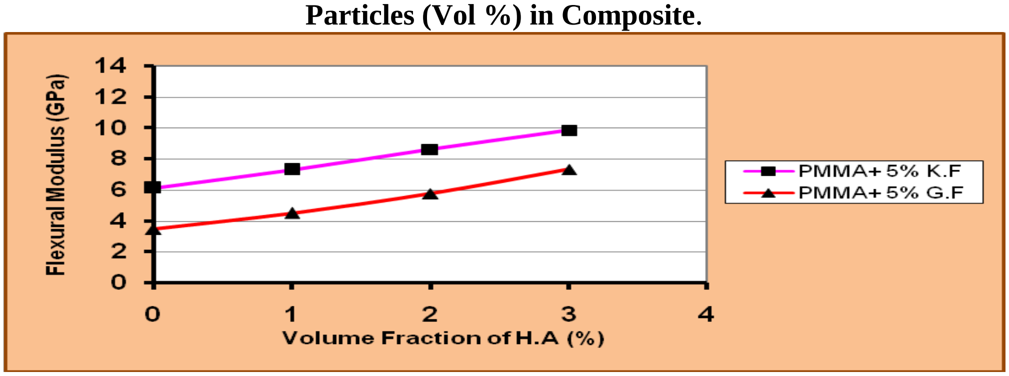
(Fig. 2): AFM Test of HA Particles.

Pure PMMA | **PMMA + nHA** | **PMMA + ZrO_2**

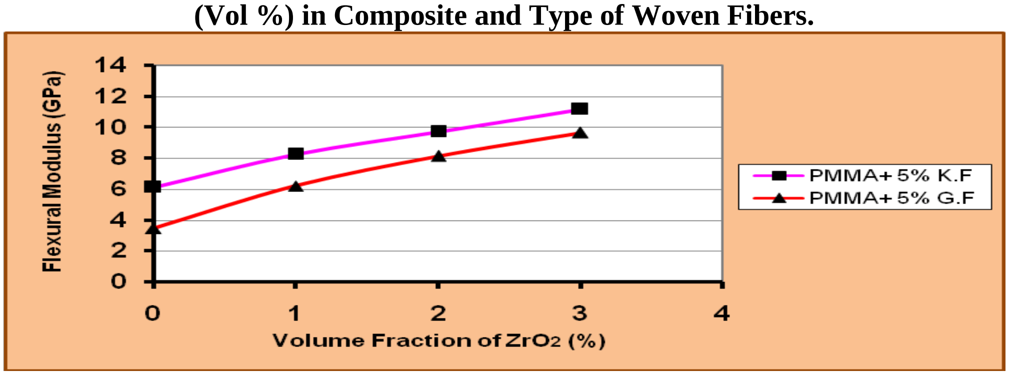
(Fig. 3 a, b and c): Showing the Casting Specimens for Pure PMMA and PMMA Composite Reinforced by (nHA or ZrO_2) Particles.



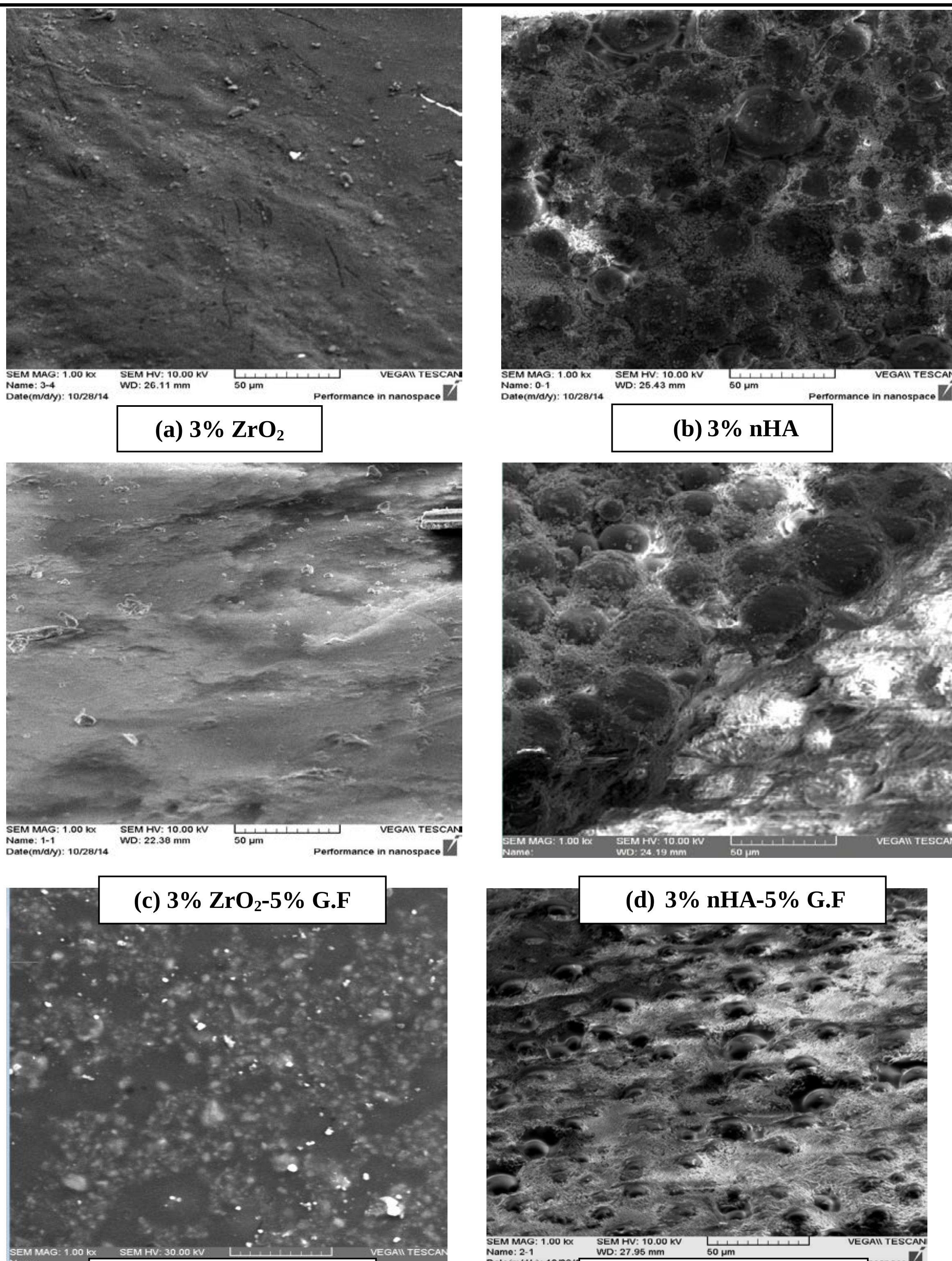
(Fig. 4): Flexural Modulus of PMMA Composite Materials as Function of (HA or ZrO_2) Particles (Vol %) in Composite.



(Fig. 5): Flexural Modulus of Hybrid Laminated Composite Materials as Function of HA Particles (Vol %) in Composite and Type of Woven Fibers.



(Fig. 6): Flexural Modulus of Hybrid Laminated Composite Materials as Function of ZrO_2 Particles (Vol %) in Composite and Type of Woven Fibers.



(a) 3% ZrO_2

(b) 3% nHA

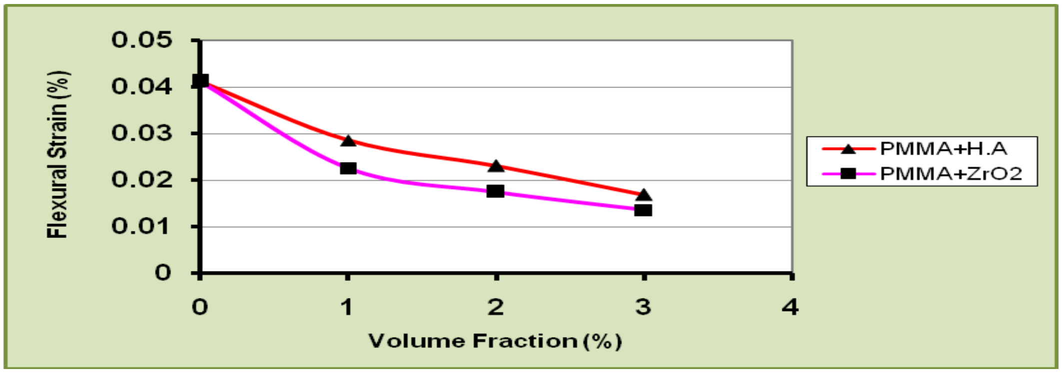
(c) 3% ZrO_2-5% G.F

(d) 3% nHA-5% G.F

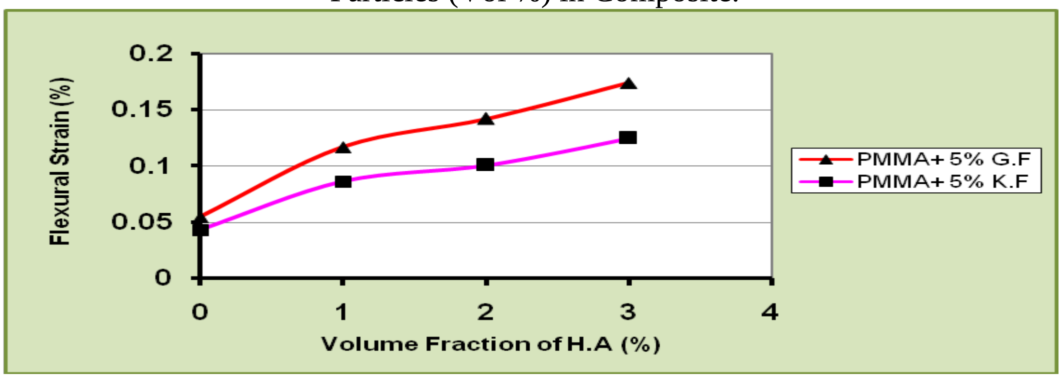
(e) 3% ZrO_2-5% K.F

(f) 3% nHA-5% K.F

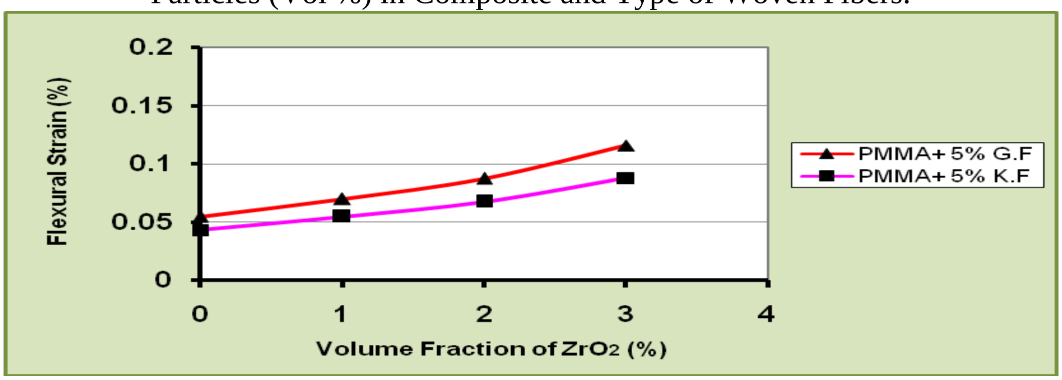
(Fig. 7 a, b, c, d, e and f): SEM Images Showing the Different in the Surface Morphology between the (PMMA-nHA) and (PMMA-ZrO_2) Composites at Magnification (1000×).



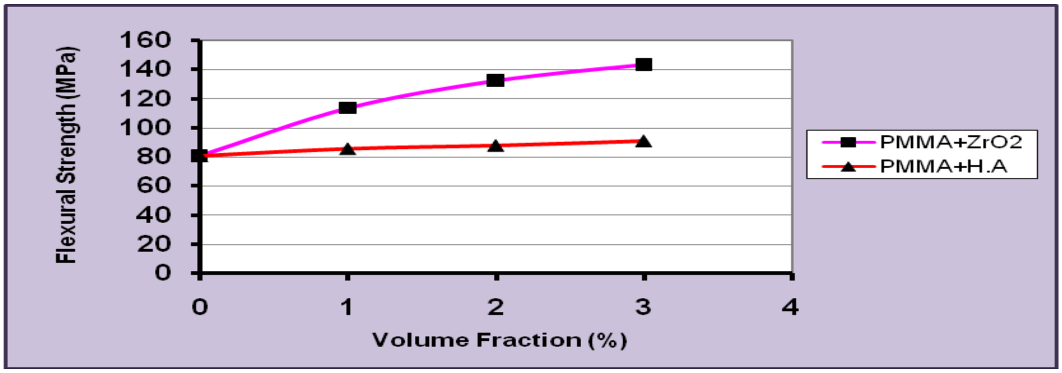
(Fig. 8): Flexural Strain of PMMA Composite Materials as Function of (HA or ZrO_2) Particles (Vol %) in Composite.



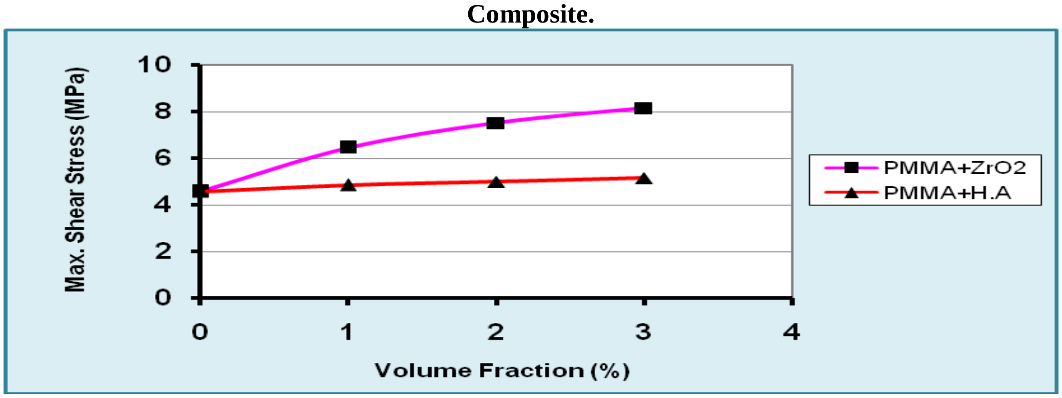
(Fig. 9): Flexural Strain of Hybrid Laminated Composite Materials as Function of HA Particles (Vol %) in Composite and Type of Woven Fibers.



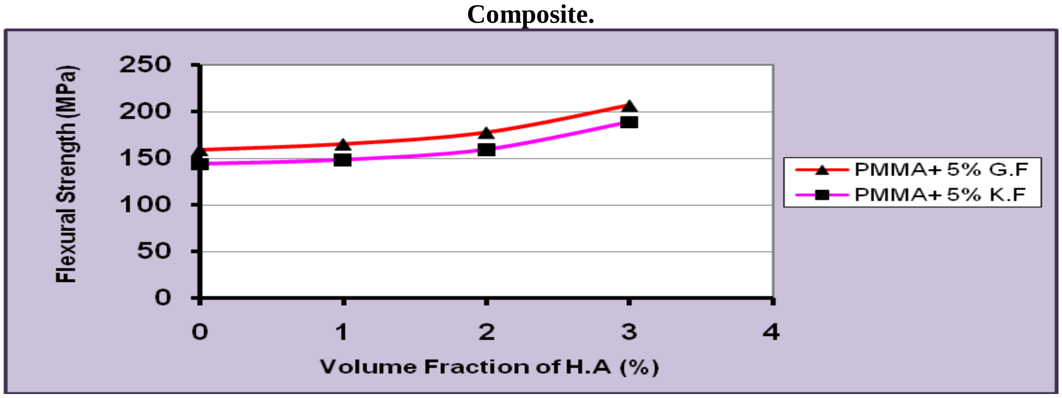
(Fig. 10): Flexural Strain of Hybrid Laminated Composite Materials as Function of ZrO_2 Particles (Vol %) in Composite and Type of Woven Fibers.



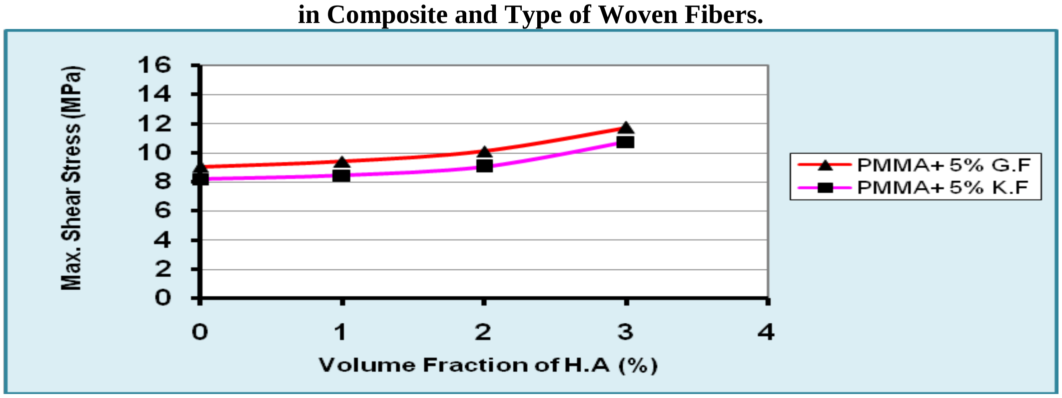
(Fig. 11): Flexural Strength of PMMA Composite Materials as Function of (HA or ZrO_2) Particles (Vol %) in Composite.



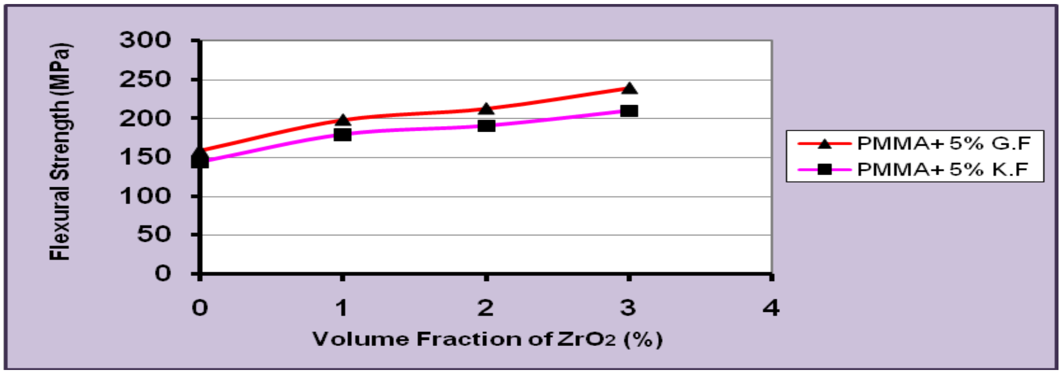
(Fig. 12): Max. Shear Stress of PMMA Composite Materials as Function of (HA or ZrO_2) Particles (Vol %) in Composite.



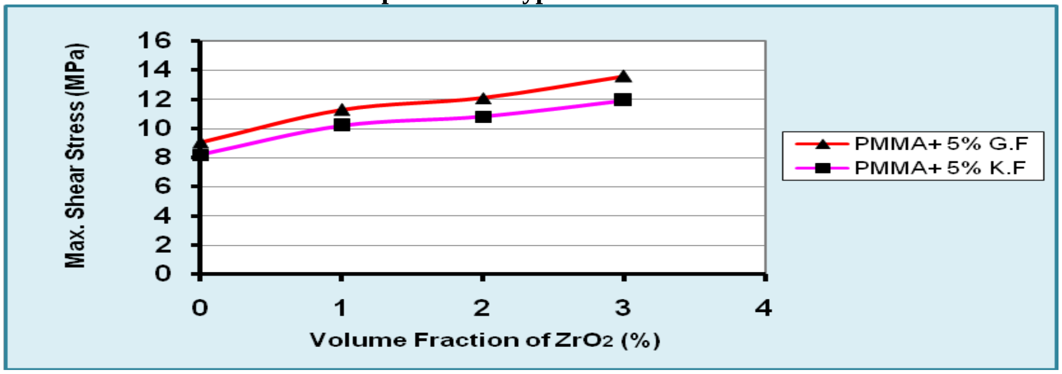
(Fig. 13): Flexural Strength of Hybrid Laminated Composite Materials as Function of HA Particles (Vol %) in Composite and Type of Woven Fibers.



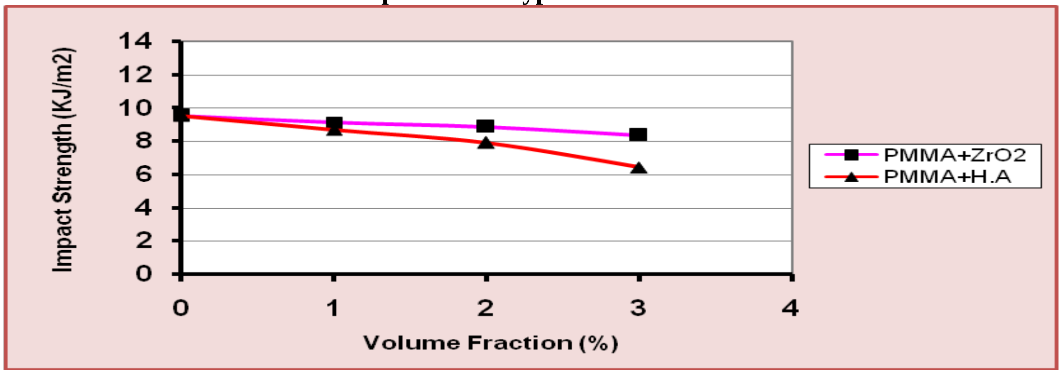
(Fig. 14): Max. Shear Stress of Hybrid Laminated Composite Materials as Function of HA Particles (Vol %) in Composite and Type of Woven Fibers.



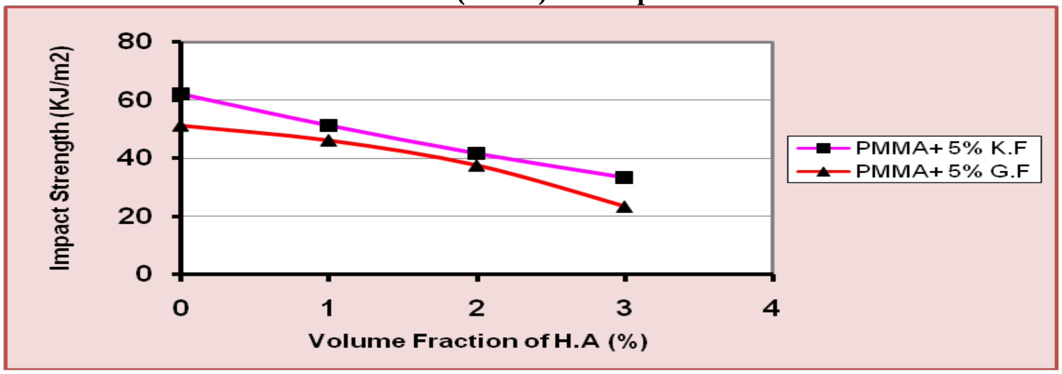
(Fig. 15): Flexural Strength of Hybrid Laminated Composite Materials as Function of ZrO_2 Particles (Vol %) in Composite and Type of Woven Fibers.



(Fig. 16): Max. Shear Stress of Hybrid Laminated Composite Materials as Function of ZrO_2 Particles (Vol %) in Composite and Type of Woven Fibers.

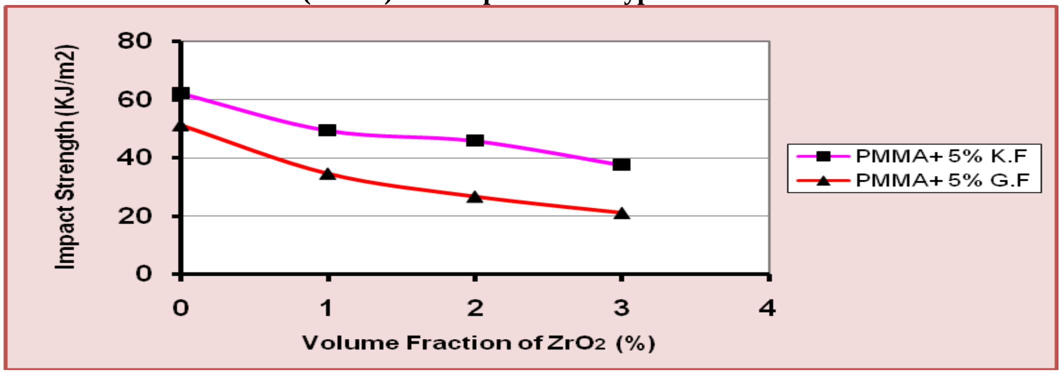


(Fig. 17): Impact Strength of PMMA Composite Materials as Function of (HA or ZrO_2) Particles (Vol %) in Composite.



(Fig. 18): Impact Strength of Hybrid Laminated Composite Materials as Function of HA

Particles (Vol %) in Composite and Type of Woven Fibers.



(Fig. 19): Impact Strength of Hybrid Laminated Composite Materials as Function of ZrO_2 Particles (Vol %) in Composite and Type of Woven Fibers.

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