ISSN No: 2348-4845
International Journal & Magazine of Engineering, Technology, Management and Research
A Peer Reviewed Open Access International Journal

Preparation and Characterization the Flexural Properties and Impact Strength of PMMA Nano Composite Used For Denture Base

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Abstract:

In the present work, it was preparation of polymer matrix Nano composite materials by used the hand lay-out technique, where the composite material was prepared from two sets of composites materials, the first set consists of a poly methyl Methacrylate (PMMA) resin as matrix material reinforced by nanoparticle of aluminum Oxide (Al_2O_3) with average size (53.60) nm once and the second group is composed of poly methyl Methacrylate resin reinforced by nanoparticle of Magnesium Oxide (MgO) with average size (52.54)nm . The research includes the study of the influence of the selected volumetric fraction of (0.5, 1, 1.5%) nanoparticlesreinforce materials, on the properties of the prepared composite materials. Number of mechanical tests, (flexural strength and Impact strength) was performed on the two groups of Nano composite, and all of these tests were done at room temperature. Result of the work shows that the values of flexural strength, flexural modulus and fracture toughness increase with the increase of volume fraction of nanoparticle for both groups of polymeric Nano composites material. Whereas decreased the values of impact with the increase of volume fraction of nanoparticle for both groups of polymeric Nano composites material. Moreover, the poly methyl methacrylate Nano composite reinforced with aluminum oxide nanoparticles, possesses higher values of flexural strength, flexural modulus and fracture toughness as compared with their counterparts of other group of PMMA Nano composites reinforced with the magnesium oxide nanoparticles.

Key words:

PMMA , nanoparticles Composite , Aluminum oxide , Magnesium oxide, flexural strength and Impact strength.

Introduction:

Denture Base is defined as the part of Denture that rests on the foundation tissue and to which teeth are attached. Denture Base Material is any substance of which denture base may be made. Acrylic prosthetic resins are used in a number of types of dental prostheses, including complete or removable partial dentures, transitional prostheses, and implant-supported prostheses. Poly (methyl methacrylate) (PMMA) resin has been widely used as a denture base material due to its desirable properties of excellent aesthetics, accurate fit, stability in the oral environment, low water sorption and solubility, relative lack of toxicity, ability to repair, dimensional stability, and simple processing techniques, have made the (PMMA) as preferred denture base material. Despite of the popularity and satisfactory esthetic properties of (PMMA), has mechanical properties inadequate as denture base materials, So, the fracture resistance of PMMA is not satisfactory [1]. Denture fracture is a multifactorial phenomenon, and even strengthening measures could not efficiently prevent denture fracture [2]. Poor fit of the denture, non-balanced occlusion, and excessive biting force are possible causes for such deficiency [3] Moreover, the failure of the acrylic resin dentures intra-orally often occurs by a fatigue mechanism where a relatively low magnitude flexural stresses over a period of time eventually lead to the formation of a small crack. This crack propagates through the denture resulting in denture fracture [4, 5]. Additionally, the poor impact resistance of this material leads to its accidental damage. the flexural fatigue and impact fracture have been implicated as a mechanism of

Figure (1) Dentures made of poly methyl methacrylate.

Aim of this search: Poly (methyl methacrylate) (PMMA) resin has been widely used as a denture base material, the mechanical properties it has inadequate as denture base materials, the fracture resistance of PMMA is not satisfactory. So, this search is an attempt to improve the flexural properties and impact strength of prepares PMMA Nano composite.

Materials and Experimental Work

Materials Used

The different materials used in this study to prepare test specimens of the Nano PMMA composites prosthetic dentures which included the following:

1. PMMA (cold cure as pour type denture base material).
2. Aluminum oxide nanoparticles(Al_2O_3).
3. Magnesium oxide nanoparticles (MgO).

The particle size distribution of these powders was carried out by Atomic Force Microscopy (AFM) in Baghdad University Laboratories by using Scanning Probe Microscopy (SPM). The result of particle size distribution of(Al_2O_3) particles is shown in Figure (2a), where the average value of diameter was (53.60nm). While the result of particle size distribution of (MgO) nanoparticles is shown in Figure (2b), and the average value diameter was (52.54nm).

Mould Preparation:

The mould used in this study is made of glass mould with dimensions (25cm × 25cm × 0.5cm) and covered with a glass plate to provide smooth surface of prepared specimens. Thermal silicon was used to close any spaces that may be found in the mould.

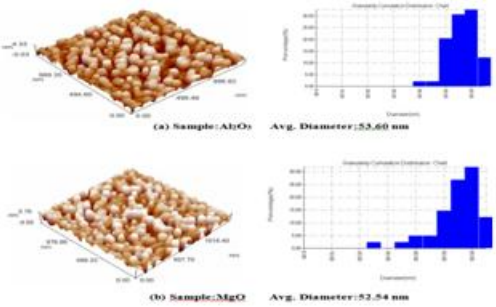

Figure (2): AFM Test of Nano (a) Al_2O_3(b) MgO

Mould Preparation

The mould used in this study is made of glass mould with dimensions (25cm × 25cm × 0.5cm) and covered with a glass plate to provide smooth surface of prepared specimens. Thermal silicon was used to close any spaces that may be found in the mould.

Preparation Method

Many methods are used to prepare the polymer matrix Nano composite materials. The (Hand lay-Up) method was used in this study. The steps of prepared specimens are shown as follows:

1) Weighting of the reinforced nanoparticles (Al_2O_3andMgO) with the selected volumetric fraction of (0.5, 1, 1.5%)

2)Weight of the acrylic resin powder (PMMA) and monomer liquid (MMA) resin. This weight was done according to the manufacturer's instructions of Manufacturer Company. which was calculated according the volume fraction of each component in composite, moreover, the requisite of casting mold, and depending on the volume fraction of reinforced material (Al_2O_3 and MgO) which was mentioned in point (1) above.

3) Mixing the mixture which consist of acrylic powder PMMA and (Al_2O_3 and MgO), with MMA liquid resin according to the steps mixing of acrylic until reach to dough mixture stage that explain priory, with different volume fraction according to specimen group. Mixture was done at room temperature, this process done by using metal spatula to ensure, that the mixture, was be done against the container wall and in order to remove any entrapped of air gas from specimen.

4) The Vertex Castavaria is moldable 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 (brabenderplastograph 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.

Mechanical and Physical Tests

Mechanical

The flexural test is performed according to (ASTM D790) [12] All data measured from three point bending test machine (model WDW 200 E) made in China, at room temperature and after fixing the ends of the sample on the supports of the instrument, the load were increased gradually on at the middle of the sample with velocity (across head speed) (5mm/min) and load was applied equal (5 kN) until the failure of the specimen occurred. The value of fracture load (F) is read from the gage. Four specimens where used for most tests and final results represent the average for four specimens it was tested. The flexural strength and flexural modulus are valued from the relationships (1 and 2) respectively [13].

$$\text{Flexural strength for three -point bend test} = \sigma_{bend} = \frac{3FL}{2wh^2}$$

…….1

where F is the fracture load, L is the distance between the two outer points, w is the width of the specimen, and h is the thickness of the specimen.

$$\text{Flexural modulus } (E_f) = \frac{FL^3}{4\partial wh^3} = \frac{mL^3}{4wh^3} \quad \text{.....2}$$

Where

∂: is the deflection of the beam when a force F is applied,
M: is the slope of the load (F) /deflection curve.

Izodcharpy Impact test was performed at room temperature according to ASTM ISO 179, the impact test instrument model XJU-22, supplied from Time group Inc, Nano research center, University of technology. Impact strengthcan be calculated from the following relationship [14]:

$$G_c = \frac{U_c}{A} \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots .3$$

Where:-

G_c : - The impact strength of the material (J/m2).
U_c : - The required energy for sample fracture (J).
A : - the cross sectional area of the sample (m2).
Fracture toughness: - can be calculated as follows:-

$$K_c = \sqrt{G_c E_b} \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots .4$$

Where:

K_c : - Fracture toughness of the sample ($N/mm^{1/2}$).
G_c : - Impact strength of the material (J/m^2).
E_b : - Young Modulus of the material (MPa).

(FTIR) test is performed according to (ASTM E1252) [15]. By using Fourier transform infrared spectrometer, model (TENSOR 27) made in Germany, by (Bruker Optics Company). It is equipped with a room temperature DTGS detector, mid-IR source (4000 to 400) cm-1 and a KBr beam splitter.

Results and Discussions

Miscibility Tests

These tests are used for fully characterization of PMMA and itis Nano composites specimens as a function to addition the Nano- alumina (Al_2O_3) and the Nano- magnesia (MgO) particles in PMMA materials. The FTIR spectrum in the frequency range (400-4000 cm^{-1}) was used in this study.

Results and Discussions of FTIR Analysis for PMMA Modified Composites:

FTIR spectrum of neat PMMA Figure 3 shows that the asymmetric and symmetric correspond to the (C–H) stretching of methyl group (CH3) in single structural repeated unit assigned to peaks at 2992.01 and 2850.27 cm^{-1}respectively, and the bands at (1447.68 and1385.47) cm^{-1} are associated with (C–H) symmetric and asymmetric stretching modes, respectively. Furthermore C=O and C-O bands are often dominates IR spectrum and appeared at range (1500-2000 cm^{-1}) and (1000-1400) respectively, The 1239.47 cm^{-1} band is assigned to torsion of the methylene group CH2 and the peak at 1189.47cm^{-1} for the band corresponds to vibration of the ester group (C–O), while (C–C) stretching band are at (986.85 and 841.11) cm^{-1} [17 and 18], in this spectrum medium strength of C=O stretching and bending assigned at 1722.63 and 749.74 cm^{-1} respectively, while medium stretching C-O assigned at 1239.74 cm^{-1}.

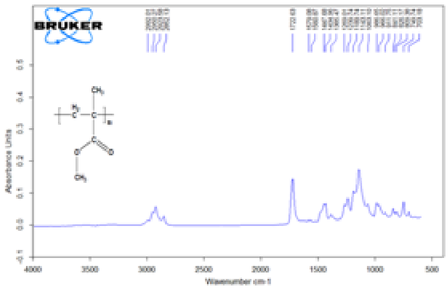

Figure (3): FTIR Spectrum that obtained for the Cold Cure PMMA Specimens.

The infrared spectrum of PMMA was reinforced with 1.5% nano-alumina as shown in Figure (4). All the characteristics vibration bands of (PMMA: 1.5% nano-alumina) composites are presented in (FTIR) spectrum. The main characteristic vibration bands (ν) of PMMA appear at 1722.23 cm-1 vibration of C = O bands and (1443.96 and1434.87) cm^{-1}ν (C– O). The bands at 2949.96 cm-1 correspond to the (C–H) stretching of the methyl group (CH3) and the bands at 1447.71 and 1385.65 cm^{-1} are associated with (C–H) symmetric and asymmetric stretching modes, respectively. The 1239.38 cm^{-1} band is assigned to torsion of the methylene group CH2 and the peak at 1189.98cm^{-1} for the band corresponds to vibration of the ester group (C–O), while (C–C) stretching band are at (986.65 and 841.22) cm^{-1} [13]. Whereas the main characteristic vibration bands (ν) of Al_2O_3 appear at 1542 cm^{-1} indicates some changes in the surface chemistry of Al_2O_3 particles. The weak peak at 1522 and 2343 cm^{-1} are assigned to nitrate and carbonate. The peak at 654 cm^{-1}, is assigned to Al-O vibrations [15]. Also, the peaks at 1635 cm^{-1} are assigned to the bending vibrations of hydroxide vibrational mode.

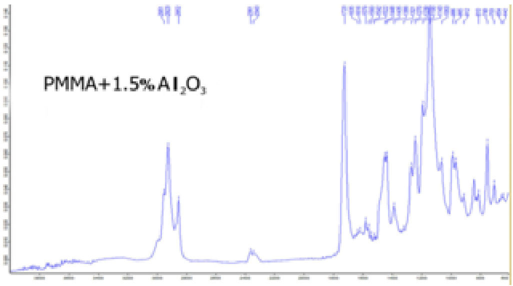

Figure 4: FTIR Spectrum that obtained for the polymer composite (PMMA: 1.5% Al2O3) Specimens.

The infrared spectrum of PMMA was reinforced with different ratio of nano- alumina particles (PMMA: x% Al_2O_3) as the first group composite materials are shown in Figure (5). All the characteristics vibration bands of PMMA composites are presented in (FTIR) spectrum of first group composites specimens.

In addition, from the infrared spectrum of the first group composite specimens as shown in Figure (5), there is no other new peak or peak shifts were observed for the PMMA composite specimens, this is due to find physical bond and absence any cross linking in these specimens. There is a clear increase in peak intensity for all of characteristic peak with increasing Nano-alumina ratio and it reaches a maximum at 1% Nano-alumina and then decreased with increased Nano-alumina ratio to 1.5%.

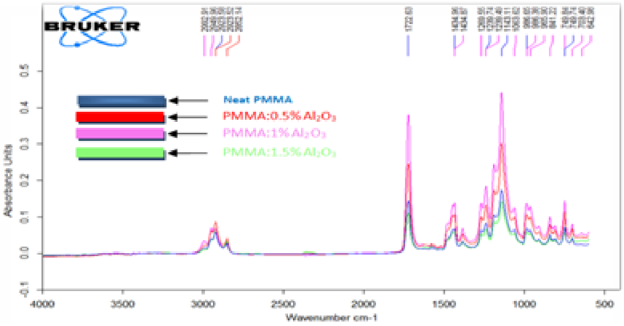

Fig (5) FTIR spectrum of (PMMA: x% Al_2O_3) polymer composite as a function of nano-alumina content in composite

The infrared spectrum of PMMA was reinforced with 1.5% nano-MgOwas shown in Figure (6). All the characteristics vibration bands of (PMMA: 1.5% MgO) composites are presented in infrared spectrum of composites specimen are similar to the FTIR spectrum of neat PMMA mentioned in (fig.(3)) , the main characteristics vibration band of PMMA appear at 1722.99 cm-1 ν (C=O) and in fingerprint at 1435.35 cm-1 ν (C–O). The presence of MgO particle in acrylic (PMMA) caused the weakening and finally the disappearance of the bands attributed to Mg–O–Mg compound is seen in the range of 487– 677 cm^{-1} as a broad band [21]. Except the peak appear at 2360 and.13 2342.32 wavenumber (cm^{-1}) are attributed to the bending vibration of absorbed water[14].

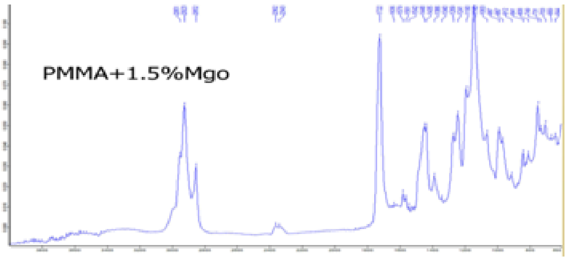

Figure (6): FTIR Spectrum for the PMMA Nano composite (PMMA:1.5% MgO) Specimens.

The infrared spectrum of PMMA and (PMMA: x% MgO) composites was reinforced with different ratio of Nano-MgO particles as the second group composite materials are shown in Figure (7). All the characteristics vibration bands of PMMA composites are presented in (FTIR) spectrum of second group composites specimens. In addition, from the infrared spectrum of the second group composite specimens (as shown in Figure (7)), there are no other new peak or peak shifts were observed for the PMMA Nano composite specimens, this is due to find physical bond and absence any cross linking in these specimens. There is a clear increase in peak intensity for all of characteristic peak with increasing MgONano-powder ratio and it reaches a maximum at 1% Nano-alumina and then decreased with increased MgONano-powder ratio to 1.5%.

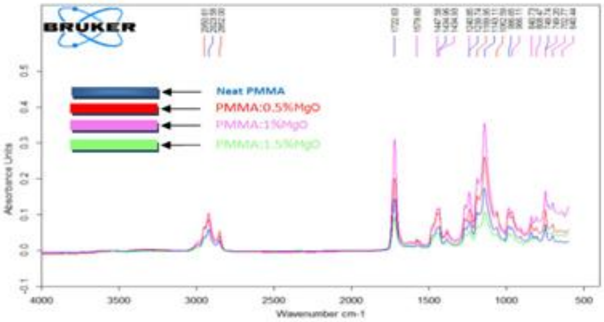

Fig (7) FTIR spectrum of (PMMA: MgO) PMMA Nano composite as a function of nano-MgO content in composite

Bending Test Results

The main objective of the flexural test is definition of the linear behavior of the specimens' materials which lie under the influence of the applied stress on the outer surface with vertical axis. Bending characteristics, which include flexural strength and flexural modulus, are shown in Figures (8 and 9) respectively. It has been noticed from these figures, that there was an increase in flexural strength and flexural modulus values with the increase in the volume fraction ratio of nanoparticle Al_2O_3 and MgO of the specimens for the first and second groups Nano composite.This is due to the ability of these nanoparticles 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 of nanoparticle Al_2O_3 and MgO are much higher than PMMA matrix. It can be noted from these figures that the addition of the zirconium oxide and MgO as nanoparticles lead to increase the flexural strength and flexural modulus, of the PMMA Nano composites as comparing with pure PMMA. The reasons behind such a behavior are that the high interfacial shear strength, between the PMMA matrix and each of zirconium oxide and MgO nanoparticles, and that related to the formation of physical cross-links bonding which shield or cover the nanoparticles and this in turn prevent the propagation of the cracks inside the material, based on the foregoing.

Moreover, the incorporation of the brittle nanoparticles into the polymer matrix improves the stiffness of the composites by restricted the mobility of the polymer chains in Nano composite. And good distribution of nanoparticle, especially at the low percentages of nanoparticles additives to the composites materials, as done in this work (the volumetric fraction of nanoparticles content in composite not heighten than 1.5% ratio) and this reduces agglomeration of the nanoparticles in composite, and that may be lead to reduces from the location and the density of stresses concentration in composite materials, so, such a small stresses are not sufficient enough to break the weak interactions at the interface. Therefore, these small stress concentrations can be easily transferred from the matrix to the brittle nanoparticles, so allowing the particles to contribute its high brittleness property to the composites so, the flexural strength and flexural modulus increases. Moreover, the formation of a strong structure of the PMMA Nano composite materials which depending on the formation of strong interfaces bonding between each of the reinforcing nanoparticles (zirconium oxide and Mgo) and PMMA matrix, so, that is resulted in forming composite materials, with strong physical bonding, therefore this required high flexural stress to break it, and this lead to increasing flexural strength and flexural modulus. It can also be noticed in these Figures that the addition of Al_2O_3 nanoparticles has a noticeable effect on the both flexural strength and flexural modulus of Nano composite specimens more than the MgO particles. Therefore, it was observed, that the flexural strength and flexural modulus for first group (Al_2O_3-PMMA) Nano composite specimens are higher in values and rate as compared with their counterpart of the other group (MgO-PMMA) Nano composite specimens. This is due to the improvement of the mechanical properties that are associated with the addition of Al_2O_3 nanoparticles which have higher hardens value as compare with MgO nanoparticles[15].

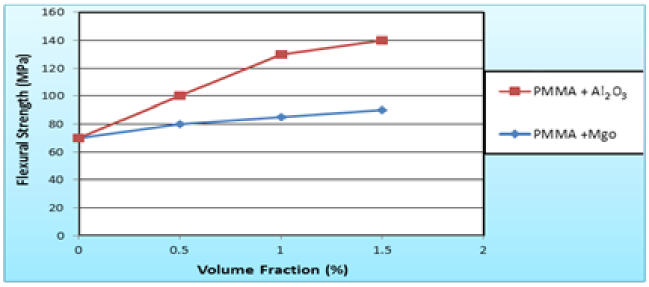

Figure (8): Flexural Strength of PMMA Composite Materials as Function of (Al_2O_3 or MgO) nanoparticles content in Composite.

ISSN No: 2348-4845
International Journal & Magazine of Engineering, Technology, Management and Research
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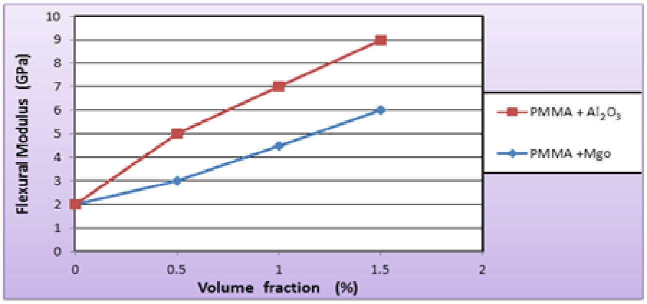

Figure (9): Flexural Modulus of PMMA Composite Materials as Function of (Al_2O_3 or MgO) nanoparticles content in Composite.

Results Impact Test

The Impact test represents one of important dynamic mechanical tests, where the specimen exposed to very fast moving load. From this test the impact strength (Gc)and fracture toughness (Kc)of composite materials can be calculated from the relationship 3 and 4 respectively. Figure (10) shows the relationship between the volume fraction of (Al_2O_3 and MgO) nanoparticles in PMMA resin and impact strength of the specimens for the first and second groups. It can be noticed that the values of impact strength decreased with increasing of the volume fraction of both types of nanoparticles for both groups Nano composite.

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. Also may be because aggregated of nanoparticles which have high surface energy that evidenced from SEM test. It can also be noticed in Figure (10) that the values of impact strength for second group (MgO-PMMA) Nano composite specimens are lower than the values of impact strength for first group (Al_2O_3-PMMA) Nano composite specimens. This is due to the higher mechanical strength such as hardness of Al_2O_3 nanoparticles comparing with MgOnanoparticles and then decreased the impact strength

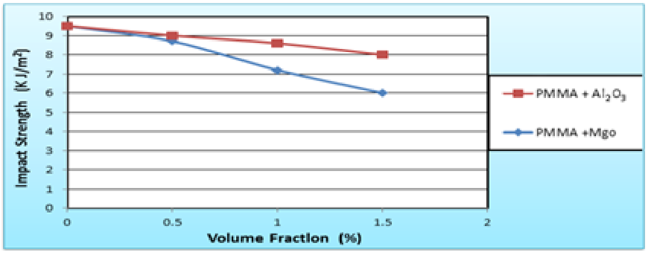

Figure (10): Impact Strength of PMMA Nano composite Materials as Functionof (Al_2O_3 or MgO) nanoparticlescontent in Composite.

While the fracture toughness appear in Figure (11) shows the relationship between the volume fraction of (Al_2O_3 and MgO)nanoparticles in PMMA resin and fracture toughness of the specimens for the first and second groups PMMA Nano composite. It can be noticed that the values of fracture toughness increased with increasing of the volume fraction of both types of nanoparticles for both groups Nano composite. This is due to the fracture toughness dependence up on the impact strength value and flexural modulus value had shown in Figure (8) and has behavior similar of the behavior fracture toughness in this Figure. Therefore, when added any type of these particles in the PMMA composite lead to hindering and obstacle the crack propagation inside the materials.

In addition to the foregoing reasons mentioned above,in item of the flexural. It can also be noticed in this Figure that the addition of Al_2O_3 nanoparticles has a noticeable effect on the fracture toughness of composite specimens more than the MgO particles. Therefore, the values of fracture toughness for first group (Al_2O_3-PMMA) Nano composite specimens are higher in values and rate as compared with values of fracture toughness of their counterpart of the other group (MgO-PMMA) Nano 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 is associated with the addition of Al_2O_3nanoparticles, that related to the nature of Al_2O_3 nanoparticles which have high hardness value comparing with MgOnanoparticles

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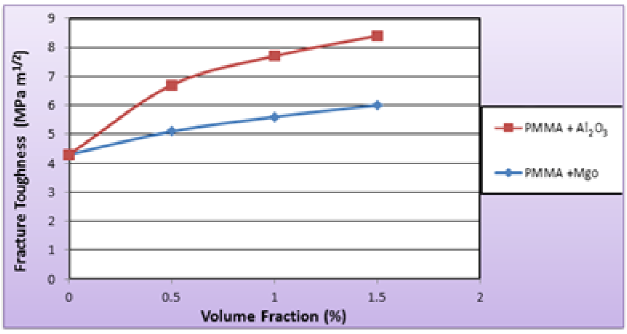

Figure (11): Fracture Toughness of PMMA Composite Materials as Function Of (Al_2O_3 or MgO) nanoparticlescontent in Composite.

Conclusions

Hence, this study was conducted to determine the strength characteristics of the Poly methyl methacrylate Nano composite used in denture base materials, in an attempt to explore the effect of addition of each of Al_2O_3 and MgO nanoparticle on some of their mechanical properties. So, it can be concluded the following:-

1. The mechanical properties of PMMA Nano composites (PMMA-Al_2O_3) and (PMMA-MgO) specimens, increased with increasing of the volume fractions of (Al_2O_3 and MgO) nanoparticles, except the (impact strength) values were decreased with increasing the volume fraction of each(Al_2O_3 and MgO)nanoparticles.
2. The addition of Al_2O_3particles has a noticeable effect on the most properties of composite prosthetic denture base materials for all groups ‘specimens as compared with their counterpart of the other group (MgO -PMMA) Nano composite specimens.
3. From infrared spectrum of PMMA Nano composite, there are no other new peaks or peak shifts were observed for PMMA Nano composite specimens, this is due to find physical bond and absence any cross linking in these specimens.

REFERENCES

1. John, J.; Gangadhar, S.A.; Shah, I. “Flexural strength of heat-polymerized polymethyl methacrylate denture resin reinforced with glass, aramid, or nylon fibers”. J. Prosthet. Dent. Vol. 86, 2001, pp 424–427
2. Darbar, U.R.; Huggett, R.; Harrison, A. “Denture fracture—A survey”. Br. Dent. J. Vol.176, 1994, pp 342–345.
3. Kim YK, Grandini S, Ames JM, Gu LS, Kim SK. “Critical review on methacrylate resin-based root canal sealers”. J Endod Vol. 36: 2010, pp 383–399.
4. N. W. Elshereksi, S. H. Mohamed1 , A. Arifin and Z. A. MohdIshak, “Effect of Filler Incorporation on the Fracture Toughness Properties of Denture Base Poly(Methyl Methacrylate)” Journal of Physical Science, Vol. 20(2), pp1–12, 2009
5. 5.Sahar Abdul-RazaqNaji, “A clinical Study of Complete and Partial Denture Fracture at Four Hospitals in Iraq “MDJ, Vol.:9 No.:2 2012 pp 201-207
6. Vallittu, P.K.; Lassila, V.P.; LappalainenNiom, R. “The effect of notch shape and self-cured acrylic resin repair on the fatigue resistance of an acrylic resin denture base”. J. Oral Rehabil. Vol. 23, 1996, pp108–113.
7. Nejatian, T.; Johnson, A.; van Noort, R. “Reinforcement of denture base resin”. Adv. Sci. Technol. Vol. 49, 2006, pp 124–129.
8. Zappini G, Kammann A, Wachter W. “Comparison of fracture tests of denture base materials”. J Prosthet Dent, Vol. 90, 2003; pp:578-85
9. Kim, S.H.; Watts, D.C. “The effect of reinforcement with woven E-glass fibers on the impact strength of complete dentures fabricated with high-impact acrylic resin”. J. Prosthet. Dent, Vol. 91. 2004, pp 274–280.
10. Lamb, D.J.; Ellis, B.; van Noort, R. “The fracture topography of acrylic dentures broken in service”. Biomaterials, Vol. 6, 1985 pp110–112.

11. Ganzarolli, S.M.; de Mello, J.A.; Shinkai, R.S.; del BelCury, A.A. “Internal adaptation and some physical properties of methacrylate-based denture base resins polymerized by different techniques”. J. Biomed. Mater. Res. Part B Appl. Biomater. Vol. 82, 2007, pp169–173
12. Annual Book of ASTM Standard, "Standard Test Methods for Flexural Properties of Unreinforced and Reinforced Plastics and Electrical Insulating Materials", D 790-03, PP. (1-11), (2003).
13. Internet, http://www.thejcdp.com/article-submission/Attachment-B.pdf, "Effect of Different Surface Treatments on the Shear and Flexural Rebond Strengths of a Micro-hybrid Composite", (2010).
14. Annual Book of ISO Standard, "Standard Test Method for UnnotchedIzod Impact Testing of Plastics", ISO-180, PP. (1-2), (2006).
15. Annual Book of ASTM Standard, "Standard Practice for General Techniques for Obtaining Infrared Spectra for Qualitative Analysis", E 1252-98, PP. (1-11), (2002).