



ISSN: 0067-2904

Effect of Additives on Impact Strength of Denture Base Resin

Wafaa A. Hussain, Furqan Salim Hashim

Department of Applied Sciences, University of Technology, Baghdad, Iraq.

Abstract

This research has studied the effect of addition glass fibers (woven and chopped) and Zirconium oxide Nano-particles (ZrO_2) with different weight percent to the conventional poly (methyl methacrylate) (PMMA). The prepared Nano-crystalline ZrO_2 powder with particle size of about 95nm was synthesized directly by sol-gel method. The gel dried at $100^\circ C$ for 1 hour and annealed at $400^\circ C$ for 3 hours.

The conventional acrylic resin prepared with 2:1 powder to liquid ratio to prepare pure sample, composite samples prepared by reinforcing PMMA with woven or chopped glass fiber (8, 12) wt.%, and reinforcing by (1,2,3) wt.% of prepared ZrO_2 Nano-powder.

The structural tests include: (XRD, AFM, and FTIR). The crystallized phase composition of dried ZrO_2 powder after annealing in air at $400^\circ C$ has been identified by x-ray diffractometry (XRD). The grain size of dried ZrO_2 Nano-particles by atomic force microscope (AFM).

The impact strength (I.S.) was measured by using Charpy impact test. The mechanical test done in order to compare the impact strength between pure and composite samples. From the result, the reinforcing PMMA with glass fiber in two forms (woven and chopped) improve the impact strength, while increasing of ZrO_2 Nano powder loading led to decrease the impact strength.

Keywords: Denture base, glass fiber, ZrO_2 Nano-powder, sol-gel.

تأثير الاضافات على مقاومة الصدمة لقاعدة راتنج طقم الاسنان

وفاء عبد الخالق حسين، فرقان سالم هاشم

قسم العلوم التطبيقية، الجامعة التكنولوجية، بغداد، العراق.

الخلاصة

في هذا البحث تم دراسة تأثير اضافة الياق الزجاج (حصيره، ومقطعه) و مسحوق الزركونيا النانوي بنسب وزنية مختلفة الى بوليمر البولي مثيل ميثاكرليت PMMA. مسحوق الزركونيا النانوي المحضر بحجم 95 نانومتر تم تحضيره مباشرة بطريقة ال sol-gel. تم تجفيف الجل بدرجة 100 سيليزي لمدة ساعة واحدة و حرقت بدرجة 400 سيليزي لمدة (3) ساعات
تم استخدام راتنج الاكرليك التقليدي بنسبه 1:2 المسحوق الى السائل لتحضير العينه النقيه، وصُنعت النماذج المترابطة بتقوية البولي مثيل ميثاكرليت بحصيرة والباف الزجاج المقطعة بنسب وزنية (8, 12) وكذلك باضافة (3,2,1) نسبة وزنية من مسحوق الزركونيا النانوي المحضر.

تتضمن الفحوصات التركيبية: (FTIR, AFM, XRD). حيث تم فحص تركيب الطور البلوري لمسحوق لزركونيا المجفف بعد تليدها بدرجة 400 سيليزي بواسطة جهاز ال (XRD). والحجم الحبيبي لمسحوق الزركونيا النانوي المجفف تم ايجاده بواسطة (AFM).
تم قياس متانه الصدمة باستخدام فحص جاري (charpy) بدون حز، تم اجراء الفحص الميكانيكي لمقارنة مقاومة الصدمة بين النماذج النقيه و المتراكبه. من النتائج، وجد ان تدعيم بوليمر ال PMMA باللياف الزجاج بالشكلين (الحصيرة والمقطعة) يحسن من متانة الصدمة، بينما زيادة مسحوق الزركونيا النانوي يؤدي الى نقصان متانة الصدمة.

Introduction

Poly (methyl methacrylate) [PMMA] material used as denture base material for the first time in 1937 by Walter Wright. From that time this material became the most excellent material as denture base material and it's became so prevalent in 1940s' [1, 2].

The PMMA has some problems such as; brittleness, low modulus of elasticity, low impact strength, and low flexural strength [3], but at the same time the PMMA has many characteristics made it used as denture material such as; ease in processing, stability in the oral environment, lightweight, excellent aesthetic properties, low cost, low water absorption, and can easily re-change and repaired its shape [1, 2].

Many materials have been used to reinforce the denture such as; metal in the form of wires, plate's, or powder but it has poor aesthetic and poor adhesion between metal and acrylic resin matrix. While the rubber toughening agents have been used as strengthen material but its expensive [3]. Ceramic nano-particles powders such as, ZrO_2 have been used to reinforce the PMMA as a bio-compatible and good aesthetic materials but no significant difference was detected in impact strength. [4, 5].

There are many types of fibers that can be used as reinforcement for denture base resins such as; polyethylene fibers, aramid fibers, and carbon fibers, but the carbon adversely effects on denture aesthetic because it's black color, while the Kevlar fibers are also used to reinforce and strength the PMMA; but they are causing problems in esthetics and difficult in polishing [3].

Glass fibers are used to enhance the mechanical properties of such as the ultimate tensile strength, transverse strength, and impact strength, it have easy manipulation, and they are esthetic [6].

A sol-gel method is process that used various solutions to manufacture various materials such as; powder of ceramics or glasses, and thin films. In this process the materials shaped by mixing the precursors together by magmatic stirrer at much lower temperature than other method. The hybrid organic-inorganic materials can be synthesized by this process and that is the major advantage of sol-gel method. New engineering materials able to be facilities by combination of inorganic and organic networks, however this materials have excellent properties made them used in wide rang applications [7].

Mohamed Ashour, et al. [6] studied the mechanical properties of PMMA reinforced with different concentration of Zirconium oxide (ZrO_2) Nano-powder. The mechanical characteristics were enhanced with the addition of ZrO_2 . While Vipul Asopa, et.al [8] evaluated the effect of zirconia (ZrO_2) in the two concentration (10% and 20%) on impact strength, surface hardness and water absorption of high impact acrylic resin and compared with control group. Impact strength and surface hardness had lower values when compared to the control group.

Arun Jaikumar, et al. [9] examined the reinforcement's effect on the impact strength of the heat polymerized acrylic denture base material (PMMA). The impact strength values of PMMA were highest when reinforced with glass fiber and followed by the reinforcement with butadiene styrene. However, the conventional denture base resins show lowest values of impact strength.

Marvdasht Branch, et al. [10] synthesized of Zirconia Nanoparticles from ($ZrOCl_2 \cdot 8H_2O$) and (urea) by sol-gel method. From the result the average diameter of the synthesized ZrO_2 nanoparticle is about 20 nm and has a very narrow particle distribution.

The aim of this work is to investigate the impact strength of PMMA resin, and studding the effect of glass fiber (woven and chopped) and ZrO_2 nanoparticles addition on impact strength.

Experimental Part

1. Preparation of ZrO₂ nanoparticles:

Materials

The materials used for preparing zirconium oxide nanoparticles (ZrO₂) were zirconium oxychlorideoctahydrate (ZrOCl₂.8H₂O) (BDH Chemicals Ltd Poole England), sodium hydroxide (NaOH) (BDH Chemicals Ltd Poole England), and distilled water.

Method

To prepare ZrO₂ nanoparticles, two solutions were prepared, first precursor solution was prepared by using 10g Zirconium Oxychlorideoctahydrate (ZrOCl₂.8H₂O) in 450 ml distilled water with constant stirring for 1 hour, and the second precursor was aqueous sodium hydroxide solution by dissolving 8.05 g NaOH in 100 ml of distilled water.

Appropriate amount of NaOH solution was added drop-wise to first precursor solution until the PH of the reaction mixture equal to 7. The reaction mixture was stirred until milky solution was formed. The distilled water was used in filtering and washing the gel. The precipitates dried at 100 °C for 2 hours and thermally annealed in an air-circulating oven at 400 °C for 1 h. The resulted powder after sintering was milled until reach fine powder.

2. Preparation of PMMA and composite material

Materials

The materials used for denture base are heat cured acrylic resin poly (methylemethacrylet), monomer methylmethacrylet (PMMA, MMA, New Stetic, Veracril[®], Colombia). The mold materials are made from Stone (Zhermack[®] Technical, eite[®] stone, NAVY BLUE, Italy) and kaolin. While, woven glass fiber with (65×65) mm², chopped (5 mm) length glass fiber (Tian tan A District ,Huling Industry Zone,Jiyuan city,Henan ,China), and prepared zirconium oxide (ZrO₂) Nano-particle powder were used to reinforce the denture base material.

Method

Denture base fabricated by specially designed flask. The flask have two parts, one half of the flask filled with freshly dental stone, while the other half invested with square kaolin (65×65×3) mm³ and poured the stone to fill the flask. The flask was shacked to remove bubbles from the stone. The inner surface of each flask half were coated with Vaseline to prevent the investment material from attaching to the cast. The flask left to dry at room temperature for 2 hours, opened and cleaned form kaolin.

To improve the adhesion between glass fiber and acrylic resin matrix, a silane coupling agent (Schaumburg, IL 60193, United States) used to wet the fiber before loading into the acrylic resin matrix. A (0.3) g silane coupling agent were added to 100 ml of water and alcohol solution (50 ml for each of them). The fiber immersed into the silane mixture for 10 min, and then dried at 100 °C for 1 hour.

Sight experimental groups of denture base were prepared. The specimens were fabricated by conventional heat-polymerized acrylic resin, the powder to liquid ratio is 2:1 by weight mixed and left for 10 minutes until reach the dough stage and then kneaded to remove the voids. In group (1) (Pure PMMA) the dough was placed directly in the mold. In the Groups which contain woven GF the dough was cut into two halves, and the woven glass fiber placed between them, while the Groups with chopped GF or ZrO₂ nano-powder the reinforcement materials were mixed with polymer powder. The mold pressed by hydraulic press at 1 Ton for 3 minutes. The flask immersed in water bath at room temperature and heated to 100 °C then left at boiling temperature for 1 hour to complete the polymerization process and then cooled at room temperature. The specimens extracted from the mold as shown in Figure-1, then cut to specific dimension (65×10×3) mm³ to all tests and polished with 350, 400, and 800-grit Sic paper.



Figure-1 Specimen of PMMA as denture.

Structural tests

X-ray Diffraction (XRD)

The phase purity, the levels of crystallinity and crystal size of zirconium oxide nanopowder were examined by X-ray diffraction (XRD) (SHIMADZU 6000 X-ray diffractometer). A CuK_α tube with radiation of wavelength $\lambda=1.54060\text{\AA}$ operated at 40 kV and 30 mA was used for the generation of X-rays, scanning speed 5 deg/min, scan mode: continuous scan.

Atomic Force Microscopy (AFM)

Atomic force microscopy used to measure size, distribution, granularity, roughness, and accumulation in micro or nano level. Scanning probe microscope (CSPM-5000) was used to measure average grain size of ZrO_2 powder.

Fourier Transform-Infrared (FTIR)

Fourier transform-infrared spectroscopy (FTIR) (SHIMADZO IRFFINITY) record in wave number range of $(400-4000\text{ cm}^{-1})$ to support the XRD findings, to give evidence of ionic substitution, and differentiate between different levels of relative crystallinity. The spectra of ZrO_2 powder was obtained from 3:100 (powder to KBr) pellet.

Impact strength test

The test was done by un-notched Charpy impact test machine (Impact tester N, 43-1). The pendulum of (2 joule) is dropped from a specific height to record the zero reading. The specimen was placed horizontally on the carrier, the pendulum dropped again to impact specimen. The energy absorbed by the material is obtained by difference between these reading values. Charpy impact strength was measured in kJ/m^2 , by applying the equation:

$$\text{I.S.} = \text{Corrected Reading} / \text{A} \dots\dots\dots \text{Eq(1)}$$

Where,

I.S.: Impact strength in kJ/m^2 .

Corrected Reading: (Energy of fracture (E) – Zeroing) in Joules.

A: Test specimen area (b*d width and thickness, respectively) in square meters.

Results and discussion:

X-ray Diffraction (XRD)

The XRD patterns of the prepared ZrO_2 nanoparticle with PH=7 at $400\text{ }^\circ\text{C}$ is shown in Figure-2. Monoclinic and tetragonal ZrO_2 phases are identified from diffraction peaks. Monoclinic- ZrO_2 is the main phase shown at angles of $(32^\circ, 33.5^\circ, 45^\circ, \text{ and } 50.5^\circ)$ and tetragonal- ZrO_2 is shown at angle (30.36°) .

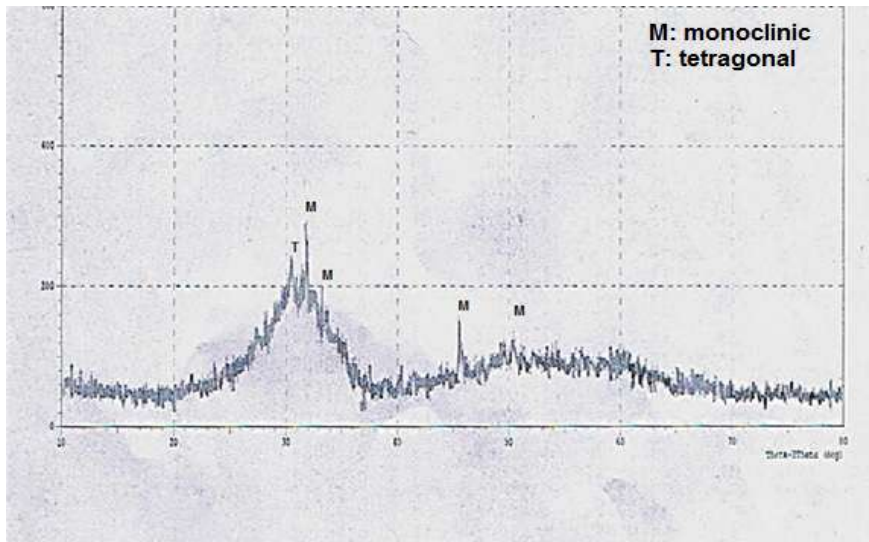
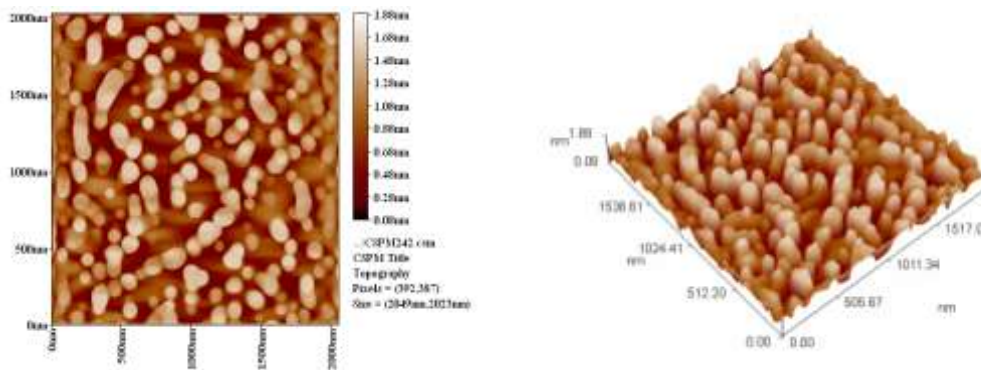


Figure 2- XRD for prepared ZrO₂

Atomic Force Microscopy (AFM)

The microstructure and surface morphology of prepared ZrO₂ powder were investigated by atomic force microscopy (AFM). Figure -3 refers to the two and three dimensional AFM images of ZrO₂ which shows that uniform distribution of ZrO₂ nanoparticles have an ellipsoidal shape with smooth surface with grain size about 95.1 nm.



2D and 3D AFM images for ZrO₂ nanoparticles

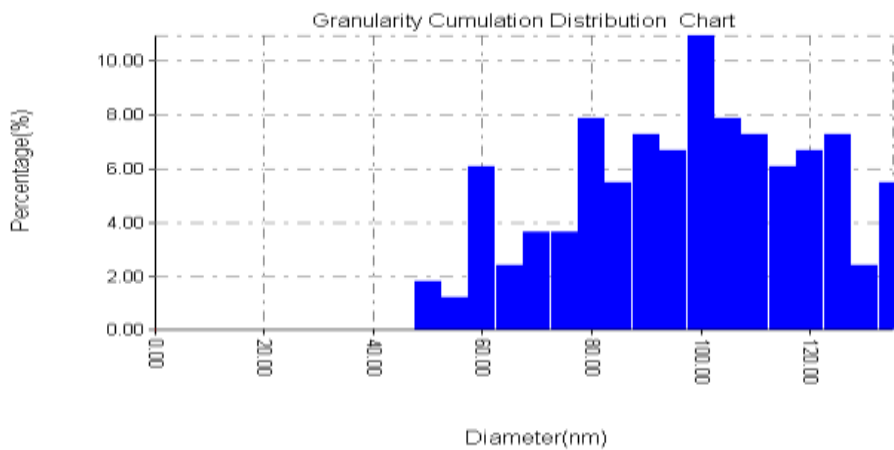


Figure 3- Granularity cumulative distribution.

Fourier Transform-Infrared (FTIR)

The FTIR spectra shown in Figure-4, the observed FTIR peak in the region of 457.14 cm^{-1} for the synthesized ZrO_2 powder is attributed to the vibration modes of ZrO_3^{-2} groups, which confirm the formation of ZrO_2 structure. prominent peak of 1346.36 cm^{-1} region refers to O=H bonding, the peaks at 1562.39 cm^{-1} and 1626.05 cm^{-1} may be correspond to adsorbed moisture (H_2O), and the wavenumber 3437.26 cm^{-1} is refers to stretching of O–H groups. The ZrO_2 is characteristic of a highly hydrated compound structure. This peaks is compatible to peaks of previous study [11].

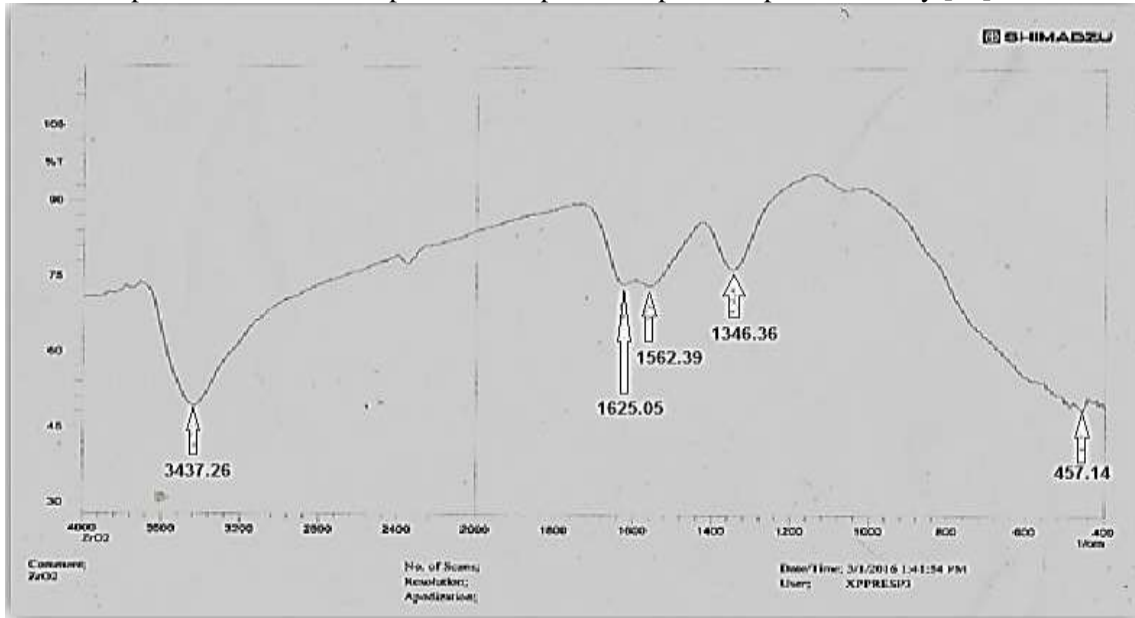


Figure 4- FTIR for prepared ZrO_2

Impact strength

The impact strength is a measure of absorb energy by the material before fracture. For denture, the impact failure can be represented by suddenly fall off dentures and collide with ground. Fracture energy for the prepared specimens is obtained from the impact test.

Heat-polymerized PMMA is a brittle material in the temperature of the oral cavity and the denture has low impact strength, to improve the impact strength for PMMA the glass fiber were used. Table (1) shows the results of impact strength for pure sample and samples reinforced with glass fiber (woven and chopped) and ZrO_2 nanoparticles. For samples prepared by woven GF, the impact strength values increased as the amount of glass fiber increases due to uniform distribution of glass fiber in cross section of sample [12, 13]. However, the incorporation of chopped GF also increased the impact strength but there is no significant change with increasing of fiber wt. % but still higher than the pure. The increment in impact strength values by using (8, 12) wt. % of woven glass fiber (G2, G3) are (50, 71.66) %, respectively, and for (8, 12) wt. % of chopped glass fiber (G4, G5) are (83.33) % as compared to control group (G1). The glass fibers had a clear improvement in impact strength when compared to unreinforced acrylic resin specimens; this could be due to proper impregnation of fibers with saline coupling agent. This finding comes in agreement with previous studies [14]. The impact strength value of sample prepared with chopped fiber is higher than sample prepared by woven fiber, because of the number of glass fiber across section area is higher in samples prepared by chopped fiber than in samples prepared by woven glass fiber. Table-1 also shows the effect of incorporation of ZrO_2 nano-powder on impact strength. The small amount of zirconium oxide shows positive effect on impact strength, while the impact strength decreased with increasing of ZrO_2 amount, in (G6) of addition of 1wt. % ZrO_2 improve the impact strength by (33%) as compared with control group (G1). This increment in impact strength is due to the interfacial shear strength between nano-filler and matrix is high, the nano-fillers in turn prevent propagation of crack. While samples with high contain of ZrO_2 (2 wt. % & 3 wt. %) (G7, G8), the impact strength did not change significantly when compared with control group, the increasing in weight percentage of nano- ZrO_2 powder affects the interface region which lead to lowering of energy dissipation per unit volume and consequently lowers the impact strength. This result is similar to previous studies [14, 15].

Table 1- the impact strength for denture reinforced with ZrO₂.

Samples		Impact strength kJ/m ²
G1	PMMA	6
G2	PMMA+ (8% woven)	9
G3	PMMA+ (12% woven)	10.3
G4	PMMA+ (8% chopped)	11
G5	PMMA+ (12% chopped)	11
G6	PMMA+ 1% ZrO ₂	8
G7	PMMA+ 2% ZrO ₂	5.7
G8	PMMA+ 3% ZrO ₂	5.4

Conclusion

1. The form of glass fiber with the same weight present affects the impact strength.
2. Volume fraction of filler ZrO₂ nanoparticles effect on impact strength.
3. It's possible to synthesis a denture base resin with good aesthetic and high impact strength by using glass fiber.

References

1. Engelmeier, R. L. **2003**. The history and development of posterior denture teeth—introduction, part II: Artificial tooth development in America through the nineteenth century. *J Prosthodont*, **12**(4): 288-301.
2. Rama, Krishna Alla1, Suresh Sajjan, Venkata Ramaraju Alluri, Kishore Ginjupalli, and Nagaraj Upadhy. **2012**. Influence of Fiber Reinforcement on the Properties of Denture Base Resins. *Journal of Biomaterials and Nanobiotechnology*, **4**: 91-97.
3. Vojdani, M. and Khaledi, AAR. **2006**. Transverse Strength of Reinforced Denture Base Resin with Metal Wire and E-Glass Fibers. *Journal of Dentistry of Tehran University of Medical Sciences*, **3**(4): 159-166.
4. Neveen, M. Ayad, Manal, F. Badawi1, and Abdou A. Fatah. **2008**. Effect of Reinforcement of High-Impact Acrylic Resin with Zirconia on Some Physical and Mechanical Properties. *Rev. Clín. Pesq. Odontol., Curitiba*, v., **4**(3): 145-151.
5. Russel, R. Ghanim. **2014**. Denture Base Modification by Reinforcement of Carbon Fiber/ Hydroxyapatite and Study its Properties. M.Sc. Thesis, Department of Applied Science, the University of Technology, Iraq.
6. Mohamed, A. Ahmed, and Mohamed, I. E. **2014**. Effect of Zirconium Oxide Nano-Fillers Addition on the Flexural Strength, Fracture Toughness, and Hardness of Heat-Polymerized Acrylic Resin. *World Journal of Nano Science and Engineering*, **4**: 50-57.
7. Podbielska, H. and A. Ulatowska-Jarza. **2005**. Sol-gel technology for biomedical engineering. *Bulletin Of The Polish Academy Of Sciences*, **53**(3): 123-131.
8. Vipul Asopa, S. Suresh, Meenakshi Khandelwal, Vivek Sharma, Shivalika S. Asopa , and Laxman Singh Kaira. **2015**. A comparative evaluation of properties of zirconia reinforced high impact acrylic resin with that of high impact acrylic resin. *The Saudi Journal for Dental Research*, **6**(2): 146-151.
9. Kaneyoshi Yoshida, Yutaka Takahashi, Ippei Hamanaka, Tomohiro Kawaguchi, Hirono Sasaki, and Hiroshi Shimizu. **2015**. Reinforcing effect of glass fiber-reinforced composite reinforcement on flexural strength at proportional limit of a repaired denture base resin. *Acta Biomaterialia Odontologica Scandinavica*, **1**(2-4): 81-85.
10. Masoud Negahdary, Amir Habibi-Tamijani, Asadollah Asadi, and Saeid Ayati. **2012**. Synthesis of Zirconia Nanoparticles and Their Ameliorative Roles as Additives Concrete Structures. *Journal of Chemistry*, **2013**, Article ID 314862, 7 pages.
11. Singh, K. and Umesh T. Nakate. **2014**. Microwave Synthesis, Characterization and Photoluminescence Properties of Nanocrystalline Zirconia. *The Scientific World Journal*, **2014**(1): 349457

12. Uciana Goguta, Liviu Marsavina, Dorin Bratu, and Florin Topala. **2006**. Impact Strength of Acrylic Heat Curing Denture Base Resin Reinforced with E-Glass Fibers. *Timisoara Medeical Journal (TMJ)*, **56**(1): 88-92.
13. Wafaa A. Hussain, and Wewench N. Rafiq. **2012**. Mechanical Properties of Carrot Fiber – Epoxy Composite. *Baghdad Science Journal*, **9**(2): 335-340.
14. Neveen M. Ayad, Manal F. Badawi, and Abdou A. Fatah. **2008**. Effect Of Reinforcement Of High-Impact Acrylic Resin With Zirconia On Some Physical And Mechanical Properties. *Rev. Clin. Pesq. Odontol., Curitiba*, **4**(3): 145-151.
15. Hussein Karim Hameed, and Hanan Abdul Rahman. **2015**. The effect of addition nano particle ZrO_2 on some properties of autoclave processed heat cure acrylic denture base material. *J Bagh Coll. Dentistry*, **27**(1): 32-39.