



## Effect of Hydroxyapatite Nanoparticles Addition on Water Sorption and Water Solubility of Heat-Cured Acrylic Resin Material

Saif Mohanad Al-Obaidy <sup>1</sup>, Ammar Khalid Al-Noori <sup>2</sup>

<sup>1</sup> Ministry of Health/ Nineveh Health Directorate / Iraq.

<sup>2</sup> Department of Prosthodontics, College of Dentistry, Mosul University / Iraq.

### Article information

Received: June 21, 2022

Accepted: August 27, 2022

Available online: March 1, 2024

### Keywords

Water sorption,  
Water solubility,  
HA nanoparticles.

### Abstract

**Aims:** To investigate the effects of the incorporation of hydroxyapatite nanoparticles (HA NPs) with size (20 nm) at two concentrations (0.5% and 1%) on the water sorption and water solubility of heat-cured acrylic resin. **Materials and methods:** The total number of specimens was thirty which was divided into ten specimens (control, 0.5 % HA NPs, and 1 % HA NPs). The statistical analysis was done by using the SPSS program including descriptive statistics, ANOVA, and Duncan's test at  $p \leq 0.05$ . **Results:** The results demonstrated that there was a significant decrease in the water solubility and a non-significantly decrease in water sorption for the PMMA-HA nanocomposite at HA nanoparticles (0.5%) and (1%), when compared to control. **Conclusions:** the use of hydroxyapatite nanoparticles as dental fillers at 0.5% and 1% by weight decreased the water sorption and water solubility of PMMA denture base material.

### \*Correspondence:

E-mail: saifmohaned93@gmail.com

تأثير اضافة جسيمات الهيدروكسي ابيتايت النانوية على امتصاص الماء والذوبان في الماء لراتنج الاكريلي المبلر بالحرارة

### المخلص

**الأهداف:** دراسة تأثير دمج جزيئات الهيدروكسي ابيتايت النانوية بحجم (20 نانومتر) بتركيزين (0.5% و 1%) على امتصاص الماء وقابلية الذوبان في الماء لراتنج الاكريليك المعالج بالحرارة. **المواد وطرائق العمل:** كان العدد الإجمالي للعينات ثلاثين مقسمة إلى عشر عينات (مجموعة السيطرة، 0.5%، 1%) هيدروكسي ابيتايت النانوية، تم إجراء التحليل الإحصائي باستخدام برنامج الاحصاء بما في ذلك الإحصاء الوصفي، اختبار انوفا، واختبار دنكن عند  $p \leq 0.05$ . **النتائج:** أظهرت النتائج أن هناك نقصان ملحوظ في قابلية الذوبان في الماء وانخفاض غير ملحوظ في امتصاص الماء لمركب النانوي المتولد في جزيئات الهيدروكسي ابيتايت النانوية (0.5%) و (1%)، عند مقارنتها بمجموعة السيطرة. **الاستنتاجات:** أن إضافة هيدروكسي ابيتايت إلى البولي ميثيل ميثاكريليت المعالج بالحرارة له تأثير ايجابي على المركب النانوي المتولد من حيث امتصاص الماء وقابلية الذوبان في الماء لمادة قاعدة أطقم الاسنان.

## **INTRODUCTION**

Teeth loss is one of the most prevalent oral health issues, particularly among the elderly <sup>(1)</sup>. There are several therapeutic procedures available to replace lost teeth, regardless of the underlying cause <sup>(2)</sup>.

Dentures, either partial or complete, are the most common treatment options for replacing missing teeth, owing to the higher expense of dental implants <sup>(3)</sup>. Resin, ceramic, and metal are the three types of dental materials utilized in denture manufacture. Acrylic resin, also known as polymethyl methacrylate (PMMA), is one of the most often used polymeric materials for denture bases because it has superior mechanical and physical qualities to other polymers <sup>(4)</sup>. Though their properties are not perfect in every way and have disadvantages such as increased water sorption and solubility, there is a need to reinforce the denture base material to increase its physical characteristics <sup>(5)</sup>.

Different processing strategies have been presented to produce a polymer with superior physical and mechanical qualities, in addition to the use of alternative materials or the modification of denture base acrylic resin by the incorporation of reinforcement materials <sup>(6)</sup>. Unfortunately, improving some properties without jeopardizing others remains challenging <sup>(7)</sup>.

Hydroxyapatite nanoparticles will not be recognized by the body as a foreign substance since they are biocompatible <sup>(8)</sup>.

This research aimed to study the influence of hydroxyapatite nanoparticle addition at two concentrations (0.5% and 1%) on water sorption and water solubility of heat-cured acrylic resin material.

## **MATERIALS AND METHODS**

### **Sampling**

The total number of specimens was thirty divided into three groups, ten specimens for each one which were (control, 0.5 % HA NPs, and 1 % HA NPs). This study was done at the College of dentistry and Technical Institute at the University of Mosul. Approval of the study was from the Scientific Research Committee / Department of Prosthodontics / College of Dentistry (UoM.Dent / DM. L.43/21).

### **Preparation of the mold**

During the mold preparation, a conventional flaking procedure was used for full dentures. A separating medium (cold mold seal) was used and allowed to dry for the layer of plastic before putting the lower part of metal flasks filled with dental stone and combining in vibration according to the directions of the manufacturer to release the trapped air, then left to set.

Acrylic sheets were utilized to make the plastic model, which was produced using computer software (AutoCAD) and then carved with a computer-controlled laser-cutting machine. The length, width, and thickness of the

plastic models used in mold fabrication were precisely established according to the specifications needed for each test. Specimens of all groups were then stored in distilled water at 37°C for 2 days using an incubator <sup>(9)</sup>.

### **Preparation of the Specimens**

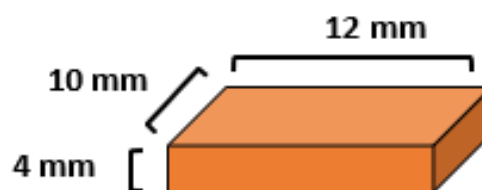
The mixing ratio of powder to liquid for heat-cured PMMA polymer material was 2:1 by weight, according to the manufacturer's instructions. The weight of the hydroxyapatite nanoparticles was subtracted from the weight of the heat-cured PMMA polymer powder to produce the precise powder-to-liquid ratio stated by the manufacturer <sup>(10)</sup>.

The specimens were first prepared by mixing the weight of hydroxyapatite nanopowder with "heat-cured PMMA" fluid monomer, which was sonicated and dispersed in the liquid monomer for 3 minutes using an ultrasonic probe of 20W and 60 kHz, and then the Heat-cured PMMA polymer powder was added and manually mixed to avoid particle agglomeration <sup>(11)</sup>.

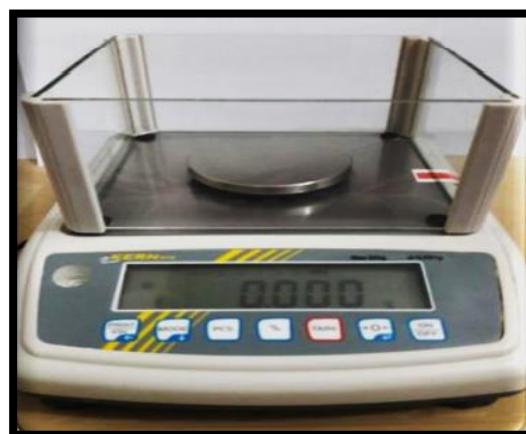
### **Water Sorption Test**

This test was carried out following ADA standard No.12 (2002). Thirty specimens were constructed, ten for each group, with the dimensions  $12 \times 10 \times 4 \pm 0.02$  mm in length, width, and height respectively (Figure 1).

**Figure (1)** Dimensions of water sorption testing specimen



After the specimens' preparation, the specimens were dried in a desiccator over silica gel at 37°C until their weight became constant. The specimens were weighed using an electronic scale with a 0.0001gm precision (Figure 2); this was their original weight ( $m_1$ ).



**Figure (2)** Electronic balance device.

Using an electronic digital caliper figure (3), the volume ( $V$ ) of each specimen was measured, and the volume ( $V$ ) was determined by taking three measurements of each dimension at three evenly spaced positions around the boundaries.

In a thermostatically controlled incubator at  $(37 \pm 1) ^\circ\text{C}$ , the samples were submerged in distilled water for 7 days  $\pm 2$  hours. After this time, the specimens were withdrawn from the water using tweezers, wiped with a clean dry towel to eliminate any apparent humidity, and weighed 1 minute later ( $m_2$ ).



**Figure (3):** Electronic digital caliper.

The samples were subsequently dried at  $37^\circ\text{C}$  in a desiccator with silica gel until they reached a constant weight ( $m_3$ ).

The following formula was used to calculate and express the water sorption ( $W_{sp}$ ) in micrograms per cubic millimeter ( $\mu\text{g}/\text{mm}^3$ ) (ADA, 2002):  $W_{sp} = m_2 - m_3 / v$   
 $m_2$ : After water immersion, the specimen's mass.

$m_3$ : Reconditioned mass of the specimen.

$v$ : Volume of the specimen.

### Water Solubility Test

Water solubility was measured using the same specimens that were used for water sorption. From the following formulae, the water solubility ( $W_{sl}$ ) value for each specimen was calculated and represented in

micrograms per cubic millimeter ( $\mu\text{g}/\text{mm}^3$ ) (ADA,2002).

$$W_{sl} = m_1 - m_3 / v$$

$m_1$ : Conditioned (dried) mass of the specimen

$m_3$ : Reconditioned mass of the specimen

$v$ : Volume of the specimen.

## RESULTS

The statistical analysis: Descriptive statistics, the test of normality, and Inference statistics (ANOVA and Duncan's test) were done by using the SPSS program version (19).

### Water Sorption Test

One-way analysis of variance (ANOVA) was used to assess the water sorption data of the control and (0.5% and 1%) hydroxyapatite nanoparticles, table (1), no substantial difference ( $P = 0.186$ ) between groups was discovered in this investigation.

**Table (1):** ANOVA for water sorption of control and HA nanoparticles groups.

SOV	SS	Df	MS	F	P
Between Groups	0.000	2	0.000	1.790	0.186
Within Groups	0.001	27	0.000		
Total	0.002	29			

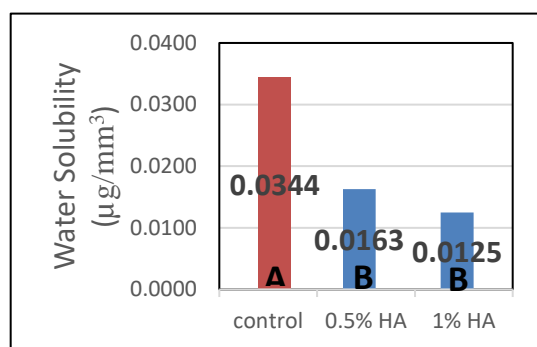
### Water Solubility Test

One-way analysis of variance (ANOVA) was used to assess the water solubility data of the control and (0.5% and 1%) hydroxyapatite nanoparticles, table (2), A substantial difference ( $P \leq 0.05$ ) between groups was discovered in this investigation.

**Table (2):** ANOVA for water solubility of control and HA nanoparticles groups.

SOV	SS	Df	MS	F	P
Between Groups	0.003	2	0.001	12.834	.000
Within Groups	0.003	7	0.000		
Total	0.006	9			

Duncan's multiple range test of water solubility demonstrated a significant decrease in HA nanoparticles (0.5%) and (1%), respectively when compared to the control. The (0.5%) and (1%) HA nanoparticle groups did not differ significantly (Figure 4).



**Figure (4):** Duncan's multiple range test for water solubility of control and HA nanoparticles groups.

## DISCUSSION

### Water Sorption

Water sorption is one of the most important aspects of denture base resin because it impacts the quality of a prosthesis and, as a result, the quality of treatment and, as a result, the patient's quality of life <sup>(12)</sup>.

The quantity of water sorption assessed in this study for both the control and experimental groups was within the

standard ADA value No. 12 (2002) for water sorption of denture base materials, which is 32 µg/mm<sup>3</sup>.

Water molecules are absorbed into the polymer due to the polarity of the polymer's molecules, the bonds of unsaturated molecules, or the polymer's imbalanced intermolecular interactions <sup>(13)</sup>.

The inclusion of hydroxyapatite nanoparticles resulted in relatively nonsubstantial differences in water sorption values when compared to the control group, as shown in Table (1).

The findings reveal that the 0.5% and 1% HA groups' water sorption values decreased but statically not significant when compared to that of the control group, this result agrees with Karadi and Hussein <sup>(14)</sup> that found the water sorption of PMMA incorporated with hydroxyapatite nanoparticles decreased but statistically not significant.

This was in agreement with Nguyen *et al.* <sup>(15)</sup> They discovered that changes in manufacturing procedures or the insertion of various additives into denture base resin might account for the observed variations in water sorption.

The degree of water sorption of polymeric materials is determined by the characteristics of the aqueous environment and the chemical structure of the polymer <sup>(16)</sup>.

Reduced water sorption might be attributed to an increase in the degree of conversion of the experimental group as well as a decrease in the amount of eluted

residual monomer <sup>(17)</sup>. Heat polymerized acrylic resins with various additives demonstrate considerable changes in the quantity of eluted residual monomer and water sorption ratios <sup>(18)</sup>.

This finding might be attributed to an increase in physical crosslinking among polymer chains or to a decrease in porosity within the resin matrix. The use of various types and concentrations of fillers minimizes acrylic resin material porosity and water absorption <sup>(19)</sup>.

### **Water Solubility**

Water solubility is one of the most essential features of denture base acrylic resin because it indicates the mass of soluble components that leak out from the polymer matrix and determines the quality of prosthesis and, as a result, the quality of patient's life <sup>(20)</sup>. The low solubility value is especially significant since the leaching of remaining monomers and/or other additives included with denture base acrylic resin, as well as their penetration into organism tissue, are both undesirable <sup>(21)</sup>.

The inclusion of hydroxyapatite nanoparticles resulted in relatively substantial differences in water solubility values when compared to the control group, as shown in tables (2).

The findings reveal that the 0.5% and 1% HA groups' water solubility values were less than that of the control group, figure (4), which might be understood by the fact that the crosslinking caused by the interlacing of the expanding polymer chains with the adjoining chain may

diminish or obstruct the possible sites for water exchange <sup>(22)</sup>.

This result was in agreement with Karadi and Hussein, <sup>(14)</sup> that found a highly significant decrease in water solubility of PMMA incorporated with hydroxyapatite nanoparticles.

The determined water solubility of the control and experimental groups was within the standard ADA value No. 12 (2002) for denture base resin water solubility, which is 1.6 µg/mm<sup>3</sup>.

The chemical structure of each kind of denture base material influences the water solubility. The incorporation of various additives into denture base acrylic resins changed their water solubility <sup>(23)</sup>. Water solubility is determined by the kind, size, and distribution of filler particles inside the resin matrix, as well as the interfacial bonding between the filler and the resin matrix <sup>(24)</sup>. This is consistent with the findings of Zidan *et al.* <sup>(25)</sup>, who discovered that the solubility of polymeric materials was dependent on the uniformity of their polymeric structure. The more homogeneous it is, the less soluble it is.

This decrease in water solubility of the experimental group could be due to increased physical crosslinking within the polymeric structure as the concentration of incorporated polymers increased, impeding the leaking of plasticizers, or to a decrease in porosity formation within the polymeric matrix which could be due to a higher degree of conversion and low content of the unreacted monomer <sup>(26)</sup>. This study's

findings agreed with those of Bacali *et al.* (27), who linked the better characteristics of acrylic resin combined with various quantities and types of fillers to a lower quantity of eluted residual monomer.

### CONCLUSION

Incorporation of hydroxyapatite nanoparticles into the "heat-cured PMMA" enhanced water sorption and water solubility of heat-cured PMMA denture base material.

### Conflicts of Interest

The authors declare that there are no conflicts of interest regarding the publication of this manuscript.

### REFERENCES

1. Peng T, Shimoe S, Fuh L, Lin C, Lin D, and Kaku M. Bonding and Thermal Cycling Performances of Two (Poly)Aryl–Ether–Ketone (PAEKs) Materials to an Acrylic Denture Base Resin. *Polymers*. 2021; 13(4):543-558.
2. Dosumu O, Ogunrinde J, and Bamigboye S. Knowledge of Consequences of Missing Teeth in Patients Attending Prosthetic Clinic. *Ann Ibd Pg Med*. 2014; 12(1): 42-48.
3. Baghani M, Yahyazadehfar N, Zamanian A, Abbasi K, Shanei F, Shidfar S, and Zohri Z. Factors Affecting Bonding Strength of Artificial Teeth: A Literature Review, *J Res Med Dent Sci*. 2018;6 (1): 184- 191.
4. Chen, S, Yang J, Jia Y, Lu B, and Ren L. A Study of 3D Printable Reinforced Composite Resin: PMMA Modified with Silver Nanoparticles Loaded Cellulose Nanocrystal. *MDPI*.2018; 11:1-14.
5. Ajay R, Suma K, and Ali A. Monomer Modifications of Denture Base Acrylic Resin: A Systematic Review and Meta-analysis. *J Pharm Biomed Sci*. 2019; 11(2): 112–125.
6. Vaishnavi A, Ganesh S, and Anjali A. Functional Modifications of Denture Base Resin - A Review. *Int J Res Pharm Sci*.2020; 11 (3): 237- 241.
7. Zafar M., Alnazzawi A, Alrahabi M, Fareed M, Najeeb S, Khurshid Z. Nanotechnology and nanomaterials in dentistry. In *Advanced Dental Biomaterials*; Khurshid, Z., Najeeb, S., Zafar, M.S., Sefat, F., Eds.; Woodhead Publishing: Cambridge, UK.2019; pp. 477–505.
8. Miljković, M, Kljajević L, Filipović S, Pavlović V.B, Nenadović S. Study of Nanosized Hydroxyapatite Material Annealing at Different Retention Times. *Sci. Sinter*. 2020; 52, 405–413.
9. Fatalla A, Tukmachi M, Jani H. Assessment of some mechanical properties of PMMA/silica/zirconia nanocomposite as a denture base material. *IOP Conference Series: Materials Science and Engineering*. 2020; 987 012031.
10. AbdulJaleel A, and Ismael T. The Effect of Aluminum Oxide Nanoparticles on Some Mechanical Properties of Room Temperature Vulcanized Maxillofacial Silicone After Artificial Aging.2019. *M.Sc. Thesis. Baghdad University, Dentistry College, Baghdad, Iraq*.

11. Lazouzi G, Vuksanović MM, Tomić NZ, Mitrić M, Petrović M, Radojević V. Optimized preparation of alumina-based fillers for tuning composite properties. *Ceramics International*. 2018;44(7):7442-9.
12. Ghasemi E, Mosharraf R, Mirzaei S. Comparison of water sorption of two injection acrylic resins with a conventional pressure-packed acrylic resin. *J Islam Dent Assoc Iran*. 2019; 31(3):177-181.
13. Narayanan, S. Comparative Study of Sorption of Heat Cure and Self Cure Acrylic Resins in Different Solution. *Int J Curr Res*. 2019;11(2):1567- 1580.
14. Karadi R, and Hussein B. Effect of modified nanohydroxyapatite fillers addition on some properties of heat cured acrylic denture base materials. *J Bagh College Dentistry*. 2017;29(2):49-54.
15. Nguyen L, Kopperud H, and Øilo M. Water Sorption and Solubility of Polyamide Denture Base Materials. *Acta Biomater Odontol Scand*. 2017; 3(1): 47–52.
16. Hamza M, Alsolam M, Oudah A, Alhadi A, and Alarjan A. Appraise the Different Types of Polymers Used in Denture Base Through their Physical Property (Water Sorption). *EIMJ*. 2020; 25(6):1-16.
17. Bacali C, Badea M, Moldovan M, Sarosi C, Nastase V, Baldea I, Chiorean R, and Constantiniuc M. The Influence of Graphene in Improvement of Physico-Mechanical Properties in PMMA Denture Base Resin. *Materials*. 2019;12(14):2335.
18. Al-Jmmal A, Mohammed N, and Taqa A. The Effect of Recycled Polymethylmethacrylate on Some Physical and Chemical Properties of Acrylic Resin Denture Base. *IJERSTE*. 2018; 7(3): 85-91.
19. Bacali C, Constantiniuc M, Moldovan M, Nastase V, Badea M, and Constantin A. Reinforcement of PMMA Denture Base Resins: From Macro to Nano Scale. *Int J Med Dent*. 2019; 23 (3): 374-378.
20. Raszewski Z, Nowakowska D, and Nowakowska-Toporowska A. The Effect of Chlorhexidine Disinfectant Gels with Anti-Discoloration Systems on Color and Mechanical Properties of PMMA Resin for Dental Applications. *Polymers*. 2021; 13(11): 1800- 1810.
21. Cierech M, Wojnarowicz J, Kolenda A, Krawczyk-Balska A, Prochwicz E, Woźniak B, Łojkowski Wm, and Mierzwińska-Nastalska, E. Zinc Oxide Nanoparticles Cytotoxicity and Release from Newly Formed PMMA–ZnO Nanocomposites Designed for Denture Bases. *Nanomaterials*. 2019; 9:13-18.
22. Taqa, A.A., Nazhat, M.N. and Basshi, T.Y. The Effect of Autoclave on the Powder of (PMMA) on the Water Sorption, Solubility and Porosity. *Int Res J Basic Clin Stud*. 2014; 2(7):87-91.
23. Mohammed, A.A. and Ismail, I.J. The Effect of Addition Plasma Treated of Polyethylene Fiber and Silanized Nanoparticles of Zirconium Oxide to Heat Cure Polyethyl methacrylate Denture Base Material on Some of its Properties. *IJSR*. 2017; 6(8) :2113-2117.
24. Dehis W, Eissa S, Elawady A, Elhotaby M.M. Impact of Nano-TiO<sub>2</sub> Particles on



- Water Sorption and Solubility in Different Denture Base Materials. *J Arab Soc Med Res.* 2018; 13:99–105.
25. Zidan S, Silikas N, Alhotan A, Haider J, Yates J. Investigating the mechanical properties of ZrO<sub>2</sub>-impregnated PMMA nanocomposite for denture-based applications. *Materials.* 2019;12, 1344.
26. Kundie, F., Azhari, C.H., Muchtar, A. and Ahmad, Z.A. Effects of Filler Size on the Mechanical Properties of Polymer-Filled Dental Composites: A Review of Recent Developments. *J Phys Sci.* 2018; 29(1), 141–165.
27. Bacali C, Buduru S, Nastase V, Craciun A, Prodan D, Constantinuc M, Badea M, Moldovan M, and Sarosi C. Solubility, Ductility and Resilience of a PMMA Denture Resin with Graphene and Silver Nanoparticles Addition. *Studia Ubb Chemia.* 2019 c;64(2):471-81.