



## EFFECT OF REINFORCEMENT OF ACRYLIC RESIN BY NANOSILVER AND NANOGOLD ON WATER SORPTION AND SOLUBILITY

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### ABSTRACT

**Purpose:** The objective of the study was to assess water sorption and solubility of acrylic denture base containing nanosilver and nanogold. **Methods:** Three concentrations of nanosilver and nanogold (0.01 %, 0.1%, 1% of polymer weight) were added to MMA monomer, cured samples were evaluated for water sorption and solubility using a method based on ISO standard. **Results:** Water sorption increased after one month aging in the groups containing the highest concentrations of nanosilver and nanogold, while solubility increased in nanosilver 0.1% and nanogold 1% groups. **Conclusion:** Water sorption and solubility increased with addition of nanosilver and nanogold depending on their concentrations.

**Key words:** Nanosilver, Nanogold, Water sorption, Solubility.

### INTRODUCTION

Poly methyl methacrylate (PMMA) has been widely used as a biomaterial in dentistry <sup>(1)</sup>. PMMA continues to be used as denture base materials because of its favorable characteristics as ease of processing, ease in repair, low cost and superior esthetics <sup>(2)</sup>. Water sorption and solubility has been recognized as problems in the dimensional stability of acrylic dentures. This condition affects the clinical performance of PMMA denture bases <sup>(3)</sup>. Al-Nori A <sup>(4)</sup> concluded that, PMMA though absorbs relatively moderate amount of water when placed in an aqueous environment, nevertheless, this water exerts significant effect on the mechanical and dimensional properties of the polymer <sup>(5)</sup>.

According to Arikan A et al <sup>(6)</sup> water sorption and solubility causes dimensional instability, thereby subjecting the material to internal stresses that may result in crack formation and, eventually, fractures of the denture <sup>(5)</sup>.

The presence of residual monomer, as a product of incomplete polymerization of material, results in

more porous structure of the material, which greatly reduces the mechanical and physical quality of the acrylic restorations and increases the absorption of liquids <sup>(7)</sup>. Testing materials for water sorption and solubility can give a better picture about decreased resiliency & increased porosity <sup>(8)</sup>.

Al-Hiloh S et al <sup>(9)</sup> used different concentrations of milled glass fibers added to PMMA and he found a significant decrease in water sorption <sup>(9)</sup>, he also concluded that addition of silanized zirconium oxide nanoparticles can significantly decrease water sorption and solubility of the modifies PMMA.

Recently nanoparticles were added to acrylic denture base materials to modify their biological, physical and mechanical properties. Nanomaterials have many unique physicochemical properties, such as ultra-small size, large surface area to mass ratio, extensive thermal stability, and high reactivity <sup>(10)</sup>. The properties of polymer nanocomposites depend on the type of incorporated nanoparticles, their size and shape, as well as the concentration and interaction with the polymer matrix <sup>(11)</sup>.

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Silver has a well-tolerated tissue response and low toxicity profile and it is more toxic than many other metals against a broad spectrum of sessile bacteria and fungi which colonize on plastic surface <sup>(12)</sup>.

Nanogold have many unique properties including biocompatibilities and surface carrier capabilities, non-cytotoxicity <sup>(13)</sup>, in addition to the extremely small size and high surface area <sup>(14)</sup>.

Chladek G et al <sup>(15)</sup> concluded that the addition of nanosilver had a significant influence on the mean sorption values of the modified acrylic resin. They found that, water sorption increased with nanosilver concentration ranged between 0.1% and 0.4%.

Arora N et al <sup>(16)</sup> concluded that addition of 0.2% AgNPs could result in a decrease in solubility of resin denture base material as compared to the unmodified material and they attributed this decrease to the hydrophobic nature of silver nanoparticles.

Azeez Z and Fatah N <sup>(17)</sup> have reported a significant decrease in water sorption and an increase in water solubility in the acrylic resin containing 0.5% nanosilver as compared to the unmodified acrylic resin.

## MATERIALS AND METHODS:

Nanosilver and nanogold were synthesized in a colloidal solution of alcohol. Nanosilver were triangular prisms in shape, while nanogold nanoparticles were spherical in shape. All nanoparticles had an average size of 20 nm. Three different concentrations of nanosilver and nanogold were used (0.01%, 0.1%, 1%) of the polymer weight.

Disk shaped metal patterns were constructed with a diameter of 50mm and thickness of 0.5mm according to ADA 1999 specification no.12 for denture base polymers <sup>(18)</sup> and were invested in flasks with dental stone. After setting of stone, the flasks were opened and the patterns were removed, leaving cavities that were used as matrixes for the fabrication of acrylic resin specimens <sup>(19)</sup>. Control and modified acrylic resin samples were made for each performed test so that (n =10) according to international organization for standardization <sup>(20)</sup>.

The samples were weighed on an AS 110/C/2 analytic scale (Radwag: Radom, Poland) with a measurement accuracy of 0.1 mg and were placed inside desiccators containing freshly dried silica gel. The desiccators were placed in a dryer at  $37 \pm 1^\circ\text{C}$ , and the samples were weighed every 24h<sup>(21)</sup>. The measurement cycles were repeated until the daily changes in mass were no higher than 0.2 mg. Stable values were registered as  $m_1$  "conditioned mass", and the samples were placed in a chamber filled with distilled water at  $37 \pm 1^\circ\text{C}$ . Three aging times were used: 1 day, 7 days and 28 days. After aging, the samples were removed from water, and all visible moisture was removed using filter paper; the samples were air-dried for approximately 15 seconds and then weighed <sup>(15)</sup>.

The registered mass was denoted  $m_2$ . The samples were placed in desiccators with freshly dried silica gel and dried until they reached a stable mass, denoted as  $m_3$ .

The sorption and solubility of each sample were calculated using the following equations:

$$2 - 3 \quad 1 \times 100\%$$

$$1 - 3 \quad 1 \times 100\%$$

where  $w$  is sorption,  $w$  is solubility,  $m_1$  is the initial mass of dried samples,  $m_2$  is the mass after aging, and  $m_3$  is the mass after the second drying step <sup>(16)</sup>.

## RESULTS

Data were statistically analyzed using F-test (ANOVA) and Post Hoc Test (LSD) A statistically significant increase in water sorption in all test groups after 1 day was reported. Only a statistically significant increase in water sorption in Nanosilver 1 % group after 1 week was reported. After 1 month the increase was statistically significant only in Nanosilver 0.1 % group and Nanogold 1% groups (table 1, figure 1).

A statistically significant increase in solubility % in all test groups of nanosilver and nanogold after 1 day was noticed. There was a statistically significant increase in solubility % in Nanosilver 0.1% group and all Nanogold groups after 1 week. After 1 month the increase was significant in Nanosilver 0.1% group and Nanogold 1% group (table 2, figure 2).

**TABLE (1)** Comparison between the studied groups according to water sorption

Water sorption	Control	Nanosilver			Nanogold			P
		0.01%	0.1%	1%	0.01%	0.1%	1%	
<b>1 day</b>								
Min. – Max.	0.96 – 1.62	1.86 – 2.81	1.82 – 2.19	1.34 – 3.03	1.92 – 3.20	1.54 – 2.85	2.27 – 3.44	<0.001*
Mean ± SD.	1.15 ± 0.24	2.33 ± 0.44	2.02 ± 0.15	2.16 ± 0.65	2.51 ± 0.52	2.15 ± 0.46	2.93 ± 0.46	
Median	1.07	2.36	2.07	2.29	2.49	2.19	2.97	
<b>P<sub>Control</sub></b>		0.001*	0.026*	0.006*	<0.001*	0.007*	<0.001*	
<b>1 Week</b>								
Min. – Max.	1.26 – 1.76	0.91 – 1.77	0.78 – 1.84	1.35 – 3.02	1.33 – 2.76	1.10 – 2.43	0.08 – 1.85	<0.001*
Mean ± SD.	1.40 ± 0.25	1.42 ± 0.30	1.51 ± 0.39	2.16 ± 0.64	2.03 ± 0.62	1.56 ± 0.48	0.67 ± 0.65	
Median	1.29	1.41	1.59	2.30	2.04	1.43	0.62	
<b>P<sub>Control</sub></b>		0.961	0.867	0.020*	0.058	0.792	0.642	
<b>1 Month</b>								
Min. – Max.	1.81 – 1.98	0.76 – 2.83	0.97 – 3.87	1.36 – 3.04	1.19 – 2.81	1.58 – 2.46	2.21 – 3.37	0.001*
Mean ± SD.	1.85 ± 0.27	1.59 ± 0.74	2.42 ± 1.0	2.17 ± 0.65	2.16 ± 0.61	2.13 ± 0.34	2.91 ± 0.46	
Median	1.37	1.48	2.30	2.31	2.22	2.24	3.05	
<b>P<sub>Control</sub></b>		0.884	0.020*	0.096	0.105	0.129	<0.001*	

*P*: p value for ANOVA test, Significance between groups was done using Post Hoc Test (Tukey)

*P<sub>Control</sub>*: p value for comparing between Control group and each other group.

**TABLE (2)** Comparison between the studied groups according to solubility %

Solubility %	Control	Nanosilver			Nanogold			P
		0.01%	0.1%	1%	0.01%	0.1%	1%	
<b>1 day</b>								
Min. – Max.	-2.20 – 0.81	1.92 – 2.93	1.84 – 2.29	0.80 – 1.75	1.57 – 2.79	1.57 – 2.79	2.39 – 3.55	<0.001*
Mean ± SD.	-1.08 ± 1.31	2.42 ± 0.45	2.11 ± 0.19	1.29 ± 0.38	2.20 ± 0.43	2.20 ± 0.43	3.05 ± 0.48	
Median	-1.65	2.42	2.18	1.18	2.26	2.26	3.06	
<b>P<sub>Control</sub></b>		<0.001*	<0.001*	<0.001*	<0.001*	<0.001*	<0.001*	
<b>1 Week</b>								
Min. – Max.	-2.10 – -1.27	4.4 – 6.53	1.57 – 2.23	0.59 – 2.0	1.33 – 2.76	1.53 – 2.67	0.02 – 4.93	0.011*
Mean ± SD.	-1.67 ± 0.35	0.72 ± 4.36	2.02 ± 0.25	1.42 ± 0.52	2.03 ± 0.62	1.98 ± 0.39	2.0 ± 1.97	
Median	-1.61	0.40	2.08	1.44	2.04	1.89	2.0	
<b>P<sub>Control</sub></b>		0.265	0.020*	0.078	0.019*	0.021*	0.020*	
<b>1 Month</b>								
Min. – Max.	-2.54 – 6.28	1.04 – 3.76	1.16 – 5.69	1.16 – 2.83	1.23 – 3.11	1.66 – 2.69	2.51 – 3.78	0.040*
Mean ± SD.	0.21 ± 3.84	2.12 ± 0.97	3.69 ± 1.99	2.0 ± 0.56	2.41 ± 0.71	2.33 ± 0.40	3.32 ± 0.47	
Median	-1.96	1.77	3.73	1.98	2.49	2.46	3.43	
<b>P<sub>Control</sub></b>		0.481	0.020*	0.556	0.317	0.359	0.050*	

*P*: p value for ANOVA test, Significance between groups was done using Post Hoc Test (Tukey).

*P<sub>Control</sub>*: p value for comparing between Control group and each other group.

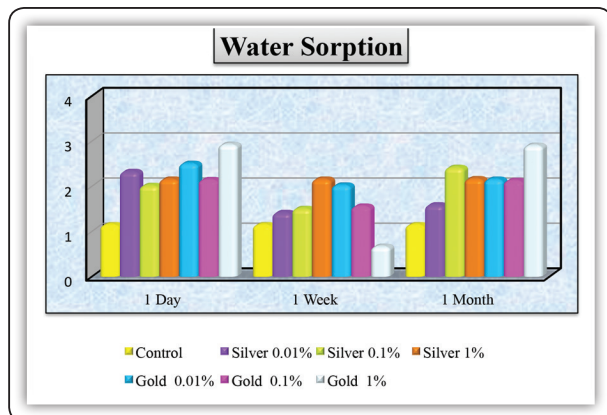


Fig. (1) Bar chart showing comparison between the studied groups according to water sorption

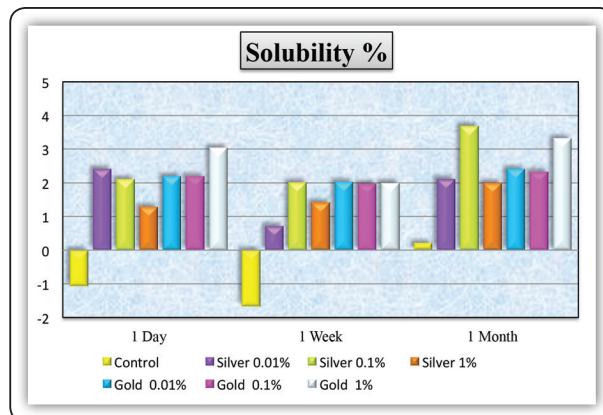


Fig. (2) Comparison between the studied groups according to solubility

## DISCUSSION

The increase of water sorption accompanying the addition of nanoparticles may be attributed to decreased cross-linking and decreased rate of polymerization which noticed by the increased time of reaching the dough stage. This time was proportional to the nanoparticles concentration<sup>(22)</sup>. The decrease in the rate of polymerization was related to the physical interaction between the nanoparticles and free radicals present in the reaction medium.

Fan C et al<sup>(23)</sup> assumed that the cross-linking problems associated with dental resin, which occur with increasing nanoparticles concentration, can be caused by the agglomeration of the nanoparticles. Based on previously reported SEM examinations of studied materials, nanoparticles agglomeration was greater in 1% AgNPs subgroup but this subgroup showed lower water sorption than AgNPs 0.1% subgroup, this could be attributed to hydrophobic nature of silver nanoparticles which became more apparent with higher concentrations<sup>(16)</sup>.

In the current study it was reported that after one month both water sorption and solubility were increased significantly in AgNPs 0.1 % and AuNPs 1% subgroups. Previous studies noticed a direct relation between water sorption and solubility, they

had found a decrease in water sorption in a modified acrylic resin material with AgNPs which was accompanying by a decrease in solubility when the measurement were done after four weeks, they based their conclusions on the facts that, water sorption and solubility were evaluated simultaneously through water gain and loss of soluble components. The resulted decrease in water sorption mean values could be attributed to addition AgNPs, with their hydrophobic nature, so addition of AgNPs resulted in decrease of micro porosity that resulted after polymerization process and subsequently reducing the water sorption. While highly significant decrease in solubility could be attributed to the decrease in water sorption properties of the modified material with the increase in the amount of AgNPs added, as indicated by their study. This limitation in the diffused water will reduce the possibility of molecular flexibility and the leach out of soluble constituents from the polymer mass<sup>(16)</sup>.

The increase in water solubility also could be attributed to the surface alteration due to nanoparticles activity and the capability to diffuse to the surface of polymer and migrate out in order to form equilibrium between the bulk and the environment<sup>(17)</sup>.

## REFERENCES

1. John J, Gangadhar S, Shah I: Flexural strength of heat-polymerized polymethyl methacrylate denture resin reinforced with glass, aramid, or nylon fibers. *J Prosthet Dent* 2001; 86:24–27.
2. Kim S and Watts D: 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* 2004; 91:74–80.
3. Tuna S, Keyf F, Gumus H, and Uzun C: The Evaluation of Water Sorption/Solubility on Various Acrylic Resins. *Eur J Dent*. 2008; 2:91–197.
4. Al-Nori A, Ali A, Rejab L: Water sorption of heat cured acrylic resin. *Al-Rafidain. Dent J* 2007;7(2):86-94.
5. Tham, W, Chow W, Mohd Isha Z: Simulated body fluid and water absorption effects on poly (methyl methacrylate)/hydroxyapatite denture base composites. *Express Polymer Letter* 2010;9: 17-28.
6. Arikan A, Ozkan Y, Arda T, Akalın B: Effect of 180 days of water storage on the transverse strength of acetal resin denture base material, *J Prosthodont*. 2010;19(1):47-51.
7. Kostić M, Petrović M, Krunić N, Igić M, Janošević P: Comparative analysis of water sorption by different acrylic materials. *Acta medica Medianae* 2104;53(2):5-9.
8. Zendeheel M, Kalateh Z, Alikhani H: Efficiency evaluation of Na Y zeolite and TiO<sub>2</sub>/ Na Y zeolite in removal of methylene blue dye from aqueous solutions. *Iran J Environ Health Sci Eng* 2011; 8: 265- 72.
9. Al-Hiloh S, Ismail I: A Study of the Effect of Addition of Silanized Zirconium Oxide Nanoparticles on Some Properties of High-Impact Heat-Cured Acrylic Resin. *J Bagh Coll Dent*. 2016;28(2):19-25.
10. Ghosh P, Han G, De M, Kim C, Rotello V: Gold nanoparticles in delivery applications. *Advanced Drug Delivery Review* 2008; 60:07–15.
11. Jordan J, Jacob K, Shart M: Experimental trends in polymer Nan composites-A review. *Material Science Engineering* 2005; 1:1-11.
12. Lok C, Ho C, Chen R, He Q, Yu W, Sun H: Silver nanoparticles: Partial oxidation and antibacterial activities. *J Biol Inorg Chem* 2007; 12:27-34.
13. Chithrani D, Dunne M, Stewart J, Allen C, Jaffray D: Cellular uptake and transport of gold nanoparticles incorporated in a liposomal carrier. *Nanomedicine: Nanotechnology and Biology Medicine* 2010; 6:61-69.
14. Khan A, Rashid R, Murtaza G, Zahra A: Gold nanoparticles: synthesis and applications in drug delivery. *Trop J Pharm Res* 2014 13; 7:69-77.
15. Chladek G, Kasperski J, Barszczewska-Rybarek I, Żmudzki J: Sorption, solubility, bond strength and hardness of denture soft lining incorporated with silver nanoparticles. *Int J Mol Sci* 2013; 14:63-74.
16. Arora N, Jain V, Chawla A, Mathur V: Effect of Addition of Sapphire (Aluminum Oxide) or silver fillers on the flexural strength, thermal diffusivity and water sorption of heat polymerized acrylic resins. *Intl. J Prosthodont* 2011;1(1):21-27.
17. Azeez Z and Fatah N: The effect of incorporation of prepared Ag-Zn Zeolite on some properties of heat polymerized acrylic denture base materials: *J Bagh Coll Dent*. 2015; 27:63–69.
18. ADA. 1999: American National Standards Institute/American Dental Association Specification No.12 for denture base polymer. Chicago: Council on Dental Material and Device.
19. Casemiro L, Martins C, Pires-de-Souza F, Panzeri H: Antimicrobial and mechanical properties of acrylic resins with incorporated silver-zinc zeolite-part I. *Gerodontology*. 2008;25(3):87-94.
20. International Organization for Standardization. 10993-5 Part 5: Test for in vitro cytotoxicity International Standard, biological evaluation of medical devices. Geneva, Switzerland: International Organization for Standardization.
21. International Organization for Standardization. 10139-2:2009: Dentistry-Materials for Removable Dentures-Part 2: Materials for Long-Term Use, 2009.
22. Soriano F, Morales G: Nanocomposites based on high impact polystyrene/silver nanoparticles: Effect of silver nanoparticles concentration on the reaction evolution, morphology, and impact strength. *Polymer Engineering and Science* 2011; 51:66–74.
23. Fan C, Chu L, Rawls H, Norling B, Cardenas H, Whang K: Development of an antimicrobial resin -A pilot study. *J Dent Mater* 2011; 27:22-28.