



Effect of Nanoparticles Incorporated to Denture Bases on Somephysicomechanicalproperties

FatheyA. ⁽¹⁾, Abd El-kawiM. ⁽¹⁾, Gaber M. ⁽¹⁾ and Fattouh D. ⁽²⁾

Codex : 09/1810

Aadj@azhar.edu.eg

KEYWORDS

*Flexural strength, stability,
thermoplastic resins*

ABSTRACT

Objective: To assess flexural strength, color stability, surface roughness of heat cured acrylic and thermoplastic resins materials after addition of (Ag &ZrO) nanoparticles.

Material and methods: One hundred specimens were prepared for this study. Fifty bars shaped specimens for flexural strength and fifty disc shaped specimens for color stability. The surface roughness was performed on the same specimens used for flexural strength. Specimens were divided into five groups according to the denture base materials, the type and concentrations of nanoparticles used. Three-point bending test was used to measure flexural strength. Color change was measured by spectrophotometer. Also roughness was assessed by using USB digital surface profile gauge.

Results: showing significant higher flexural strength of Acrylic resin than Flexible specimens that increased with increasing the AgNPs concentration led to an increase in the flexural strength of both materials than control groups, but with ZrO modified subgroup; the flexural strength decrease with increasing ZrONPs concentrations. Color showed significant changes for acrylic group than flexible group with Ag modified subgroup, but for ZrO modified subgroups, flexible group recorded significant changes than acrylic resin group. Finally, there were non-significant differences in surface roughness occurred with all groups with different nano-particle type or concentration.

Conclusion: flexural strength depends on several factors including; the type of denture base, types and concentrations of nanoparticles used. Also significant color differences were detected between control group and other tested groups that were clinically unacceptable. Surface roughness of all groups showed non-significant differences.

INTRODUCTION

Acrylic resin was the most commonly used material for construction of complete and partial denture due to their esthetic value, easy use and ability for relining and rebasing ^(1,2)

Thermoplastic material for dental prostheses, Valplast (United States) and flexible (Germany) were related to polyamides group and

1. Prosthodontics Dep., Faculty of dentistry, Al-azhar Univ., Assiut branch.
2. Prosthodontics Dep., Faculty of Dentistry, Al-azhar Univ., Cairo.

used for dental applications (Nylon Plastic) ⁽³⁻⁵⁾. Both materials used to create flexible tissue born partial denture. These materials were not strong enough to be used for conventional tooth born rest seat. The flexibility enhanced patient comfort and affect their satisfaction ^(4,6).

Many attempts have been made to enhance the strength of denture resin bases including the addition of metal wires and cast metal plates ^(7,8). The primary problem with using metal wire is poor adhesion between the wire and resin, which leads to insignificant enhancement of mechanical properties. Although metal plates increase the strength, they chemical structure, by adding crosslinking agents or copolymerization with rubber, result in significant increases in impact strength. However, stiffness, fatigue resistance, and transverse strength are reduced ^(9,10).

Mechanical reinforcement of acrylics has also been attempted through the inclusion of fibers and metal inserts ⁽¹¹⁻¹³⁾. Although the inclusion of the fibers produced encouraging results, this method has various problems including tissue irritation, increased production time, difficulties in handling, the need for precise orientation, and placement or bonding of the fibers within the resin ⁽¹³⁾. In the case of metal inserts, failure due to stress concentration around the embedded inserts has been reported ^(14,15).

The incorporation of ceramic particles in various dental materials has been studied and found to be biocompatible, and it also improves mechanical properties. In addition, the white color of the ceramic powder is not expected to compromise aesthetic appearances ⁽¹⁶⁻¹⁹⁾.

However, reinforcement methods should not have adverse effects on the mechanical properties of denture materials. The roughness of acrylic resin surfaces is a critical property because surface irregularities increase the likelihood of microorganisms remaining on the denture surface after the prosthesis is cleaned ^(20,21)

Recently, much attention have been directed toward the incorporation of inorganic nanoparticles into PMMA to improve its properties 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 ⁽²²⁾.

Addition of nano-sized particles to polymer resin denture base had been preferred to micro-sized particles because the nanoparticles characterized by better processing, smoother surfaces and larger total surface area ⁽²³⁾.

The AgNps are one of the most commonly used nano particles because of their ductility, electrical and thermal conductivity, and antimicrobial activity ⁽²⁴⁻²⁶⁾. They have shown anti-microbial effects on many microorganisms such as *E. coli*, *Staphylococcus aureus*, *Staphylococcus epidermidis*, *Candida albicans* and *Streptococcus mutans* ^(27,28). Therefore it seems that using AgNps in acrylic resins induces antimicrobial property in them ⁽²⁹⁾. Utilization of elemental silver benefits the long-term silver release in comparison to utilization of silver ions ^(30,31). In addition silver release of polymers containing silver nano particles is more effective than polymers using silver in micrometer dimensions ⁽³²⁾. As nano particles have the potential of imparting some dental materials 'mechanical properties' ⁽³³⁾, one might assume that addition of AgNps to acrylic resins affects their mechanical characteristics. Therefore, although addition of AgNps has antimicrobial advantage, we should also be concern for its effects on the PMMA flexural strength.

Adding zirconia NPs was suggested to improve the mechanical properties of PMMA ^(34,35). Incorporating zirconia NPs in PMMA increased its impact strength, flexural strength, ^(34,36,37) compressive strength, fatigue strength, as well as its fracture toughness and hardness ^(36,38,39).

The aim of this study was to assess flexural strength, color stability and surface roughness of heat cured acrylic resin and thermoplastic resin-denture base materials after addition of (Ag&ZrO) nanoparticles.



MATERIAL AND METHODS

The materials used in this study are heat cured acrylic resin (Acrostone, Acrostone England) and thermoplastic resin (iFlex by tcs, Thermoplastic comfort system, Inc. 2619 Lime Ave. signal – lill, CA90755 U.S.A) denture base materials. One hundred and fifty specimens were prepared for this study.

Flexural strength test:-

Guided with American Dental Association (ADA) specification no. 12, metallic bars were used to prepare fifty specimens of the test in the dimensions 65mm × 10mm × 3.3 mm according to specification No.12 for denture base polymers, ten specimens as control group (without nanoparticles) and forty specimens with silver and zirconium oxide nanoparticles (2% & 5%) These specimens were processed according to the manufacturer's instructions as follow:

1- Preparation of acrylic specimens containing silver nanoparticles:-

Acrylic specimens were prepared into two groups, five specimens of each, the first group was containing silver nanoparticles with concentration of 2% and the second group was containing silver nanoparticles with concentration of 5%.

Method of fabrication of specimens:

The conventional dental compression moulding technique using gypsum investment was applied to prepare specimens. Fabrication of metallic bar with dimensions (65mm × 10mm × 3.3 mm). These metallic bars were painted with separating medium, flaked in dental stone and removed from the flask after setting of the dental stone leaving mould spaces having the same dimensions of the metallic patterns. Silver nanoparticles were measured using electrical balance device of 0.0001gm accuracy and mixed with resin monomer in concentration of 2 and 5% and sonicated for 15 min in a sonicator (ELmasonic S 60 H, Germany). Afterwards, mixing

of the material in accordance with manufacturer's instructions, the polymer to monomer ratio was 3:1 by volume or 2.5:1 by weight, the material was mixed until reaching the dough stage. Packing of specimens in the moulds. Then curing by inserting the flasks in to boiling water. The specimens then were removed from mould and finished & polished. The specimens were disinfected by immersing in glutaraldehyde 2% for 2 minutes and rinsed with sterile water.

2- Preparation of acrylic specimens containing zirconium oxide nanoparticles:-

The same method as described before but zirconium oxide nanoparticles were incorporated in to the polymer powder as it is denser than silver nanoparticles. Specimens were prepared into two groups, five specimens of each, the first group was containing zirconium oxide nanoparticles with concentration of 2% and the second group was containing zirconium oxide nanoparticles with concentration of 5%.

3- Preparation of thermoplastic flexible specimens containing silver and zirconium oxide nanoparticles:

Specimens were prepared into four groups, five specimens of each, the first group was containing silver nanoparticles with 2% concentration, the second group was containing silver nanoparticles with 5% concentration, the third group was containing zirconium oxide nanoparticles with 2% concentration and the fourth group was containing zirconium oxide nanoparticles with concentration of 5%.

Method of fabrication of specimens:

Firstly, cartridges emptied from thermoplastic flexible granules were weighed using electrical balance of 0.0001gm accuracy and then the granules were put inside the cartilage and weighed. The silver and zirconium oxide nanoparticles powder (2% and 5%) of the weight of the granules were

weighed and placed a side in a plastic tube. Then the cartridge was placed on a vibrator (Vibrational Sieve Shaker, D-3162, Kottermann laborotechnik Co., Germany), and the nanoparticles powder was added gradually in to the cartridge then the cover of the cartridge was closed. The cartridge also was shaken very well manually for one minute to be ensured that the nanoparticles powder has been distributed uniformly among the granules, followed by placing the cartridge on the vibrator horizontally for two minute; the cartridge was vibrated and rolled manually to avoid accumulation of nanoparticles powder in the cartridge. The material was flasked using specially designed equipment for injection. Gypsum moulds in the flasks were obtained by investing metallic disks with the same dimensions of the acrylic specimens after painting with separating medium. The metallic disks were connected with each other and with the outside of the flask by waxed sprues. Flask was immersed in boiling water for elimination of waxed sprues. After cooling, the flask was opened and the metallic disks were removed from the flask leaving mould spaces having the same dimensions of the metallic patterns. The furnace was set in accordance with the manufacturer's instructions (230°C for 13 min) to allow the granules inside the cartridge to melt. The two halves of the flask were tightened by the 4 screws securely and placed inside the injection unit in vertical position in its correct position with the aid of the projection present at the base of the injection unit (Thermogect, Egyptian Engineering, Egy.) waiting for the granules to melt and to be injected later on. After the furnace reached the desired melting temperature and time the material was injected inside the flask. A pressure injection of nitrogen gas was used (5 Kgf) to direct the melted polymer to the areas of the mould to be filled. After 5 minutes the pressure was released, the flask is removed and allowed to bench cooling to room temperature. The flask then opened and the specimens were removed from the mould. The specimens were smoothed prior to polishing with the smooth blue rubber wheels on the mandrills and polished with pumice on a black

brush. Then specimens are disinfected by immersing in glutaraldehyde 2% for 2 minutes and rinsed with sterile water.

Conduct of the test:

All samples were individually and horizontally mounted in a custom made loading fixture [three point bend test assembly; two parallel stainless steel rods with span length 50 mm supporting the specimen, with the damage site centrally located on the tensile side] on a computer controlled materials testing machine (Model 3345; Instron Industrial Products, Norwood, MA, USA) with a loadcell of 5 kN and data were recorded using computer software (Instron® Bluehill Lite Software). Then the samples were statically compression loaded until fracture at a crosshead speed of 5 mm/min. The Stress-strain curves were recorded with computer software (Instron® Bluehill Lite Software). FS represents the limiting stress at which failure or instability is imminent. The value of the calculation of FS was guided by the formula:

$$FS (\sigma) = 3F (L) / 2wh^2$$

Where; Where; F is the maximum load at the point of fracture, L is span, w is the width of the sample and h its height.

Color stability test:

Guided with American Dental Association (ADA) specification no. 12 for denture base polymers, Hard polyvinyl plastic round discs were used to prepare both the acrylic resin and flexible specimens of the test in the dimensions 50mm in diameter and 0.5mm in thickness. These specimens were processed according to the manufacturer's instructions as described before. Fifty specimens were prepared for this test and classified into groups as described for flexural strength test.

Conduct of the test:

The specimens' colors were measured using a Reflective spectrophotometer (X-Rite, model RM200QC, Neu-Isenburg, Germany). The aperture



size was set to 4 mm and the specimens were exactly aligned with the device. A white background was selected and measurements were made according to the CIE $L^*a^*b^*$ color space relative to the CIE standard illuminant D65. The color changes (ΔE) of the specimens were evaluated using the following formula:

$$\Delta E_{\text{CIELAB}} = (\Delta L^*2 + \Delta a^*2 + \Delta b^*2)^{1/2}$$

Where: L^* = lightness (0-100), a^* = (change the color of the axis red/green) and b^* = (color variation axis yellow/blue).

Surface roughness test:

Testing of surface roughness was performed using specimens prepared as for flexural strength test.

Conduct of the test:-

Surface roughness (R_a) of each specimen of the control groups (denture base without nanoparticles) of both acrylic and flexible denture base materials was measured with USB digital surface profile gauge, (Elcometer 224/2, Elcometer Instruments, Great Britain), Figure (24) and data were recorded using computer software of roughness tester supplier (Elcomaster 2, Elcometer Instruments). For every reading made, the mean roughness value ($R_a, \mu\text{m}$) was represented by the arithmetic mean between the peaks and valleys registered, after the needle of the profilometer had scanned a stretch of 2 mm in length, with a cut-off of 0.25mm to maximize the filtering and the undulation on the surface. Each surface was read three times, always with the needle scanning the geometric center of the specimen, starting from three different points. The mean value of the three readings yielded the mean value of the roughness of each specimen. The difference in surface roughness between the control groups and other groups was calculated. The results were then tabulated and statistically analyzed.

The data will be collected, tabulated, and statistically analyzed.

RESULTS

A. Flexural strength test:

Flexural strength (MPa) results (Mean \pm SD) for both denture base materials groups as function of silver (Ag) and Zirconium oxide (Zro) nanoparticles modification and concentration are summarized in table (1).

With control subgroup; it was found that Acrylic Resin group recorded statistically significant higher mean value (59.7458 MPa) than Flexible group (17.1623 MPa) as indicated by unpaired t-test ($P < 0.0001 < 0.05$).

With 2% Ag modified subgroup; ; it was found that Acrylic Resin group recorded statistically significant higher mean value (61.0244 MPa) than Flexible group (20.1253 MPa) as indicated by unpaired t-test ($P < 0.0001 < 0.05$).

With 5% Ag modified subgroup; ; it was found that Acrylic Resin group recorded statistically significant higher mean value (66.0479 MPa) than Flexible group (23.0974 MPa) as indicated by unpaired t-test ($P < 0.0001 < 0.05$).

With 2% Zro modified subgroup; ; it was found that Acrylic Resin group recorded statistically significant higher mean value (58.2969 MPa) than Flexible group (22.9278 MPa) as indicated by unpaired t-test ($P < 0.0001 < 0.05$).

With 5% Zro modified subgroup; ; it was found that Acrylic Resin group recorded statistically significant higher mean value (54.9585MPa) than Flexible group (18.8595 MPa) as indicated by unpaired t-test ($P < 0.0001 < 0.05$).

Total effect of material type:

Regardless to nano-particle modification or concentration, totally it was found that Acrylic Resin group recorded statistically significant higher mean value than Flexible group as indicated by three-way ANOVA-test ($P < 0.0001 < 0.05$).

Table 1: Flexural strength results (Mean±SD) for both denture base materials groups as function of silver and Zirconium oxide nanoparticles modification and concentration

Control		Acrylic Resin		Flexible		Statistics
		Mean	SD	Mean	SD	P value
		59.7458	5.65	17.1623	2.13	<0.0001*
Ag modified	2%	61.0244	8.61	20.1253	1.97	<0.0001*
	5%	66.0479	11.96	23.0974	1.45	<0.0001*
ZrO modified	2%	58.2969	7.14	22.9278	1.75	<0.0001*
	5%	54.9585	7.99	18.8595	2.60	<0.0001*
Statistics	P value	0.0119*		<0.0001*		

*; significant ($p < 0.05$)

ns; non-significant ($p > 0.05$)

Total effect of nano-particle type:

Regardless to material group or nano-particle concentration, totally it was found that non-modified control subgroup recorded statistically non-significant highest mean value then Ag modified subgroup while the lowest value recorded for Zro modified subgroup as indicated by three-way ANOVA-test ($P = 0.9379 > 0.05$).

Total effect of nano-particle concentration:

Regardless to material group or nano-particle type, totally it was found that 2% modified subgroup recorded statistically non-significant higher mean value than 5% modified subgroup as indicated by three-way ANOVA-test ($P = 0.8022 > 0.05$).

B) Color stability test:

Color change (ΔE) results (Mean±SD) for both denture base materials groups as function of silver (Ag) and Zirconium oxide (Zro) nanoparticles modification and concentration are summarized in table (2).

With 2% Ag modified subgroup; it was found that Acrylic Resin group recorded statistically significant higher mean value ($18.27\Delta E$) than Flexible group ($10.76 \Delta E$) as indicated by unpaired t-test ($P = < 0.0001 < 0.05$).

With 5% Ag modified subgroup; it was found that Acrylic Resin group recorded statistically significant higher mean value ($23.19\Delta E$) than Flexible group ($13.58\Delta E$) as indicated by unpaired t-test ($P = < 0.0001 < 0.05$).

With 2% Zro modified subgroup; it was found that Flexible group recorded statistically significant higher mean value ($10.77\Delta E$) than Acrylic Resin group ($4.72\Delta E$) as indicated by unpaired t-test ($P = < 0.0001 < 0.05$).

With 5% Zro modified subgroup; it was found that Flexible group recorded statistically significant higher mean value ($11.93\Delta E$) than Acrylic Resin group ($7.17\Delta E$) as indicated by unpaired t-test ($P = < 0.0001 < 0.05$).

Total effect of material type

Regardless to nano-particle modification or concentration, totally it was found that **Acrylic Resin group** recorded statistically significant higher mean value than **Flexible group** as indicated by three-way ANOVA-test ($P = < 0.0001 < 0.05$).

Total effect of nano-particle type

Regardless to material group or nano-particle concentration, totally it was found that **Ag modified subgroup** recorded statistically significant higher mean value than **Zro modified subgroup** as indicated by three-way ANOVA-test ($P = < 0.0001 < 0.05$).



Total effect of nano-particle concentration

Regardless to material group or nano-particle type, totally it was found that 5% modified subgroup recorded statistically significant higher mean value than 2% modified subgroup as indicated by three-way ANOVA-test ($P=0.008<0.05$).

Table 2: Color results (Mean±SD) for both denture base materials groups as function of silver and Zirconium oxide nanoparticles modification and concentration

		Acrylic Resin		Flexible		Statistics
		Mean	SD	Mean	SD	P value
Ag modified	2%	18.27	0.57	10.76	1.29	<0.0001*
	5%	23.19	0.39	13.58	0.66	<0.0001*
ZrO modified	2%	4.72	0.31	10.77	0.6	<0.0001*
	5%	7.17	0.44	11.93	0.81	<0.0001*
Statistics	P value	<0.0001*		<0.0001*		

*; significant ($p<0.05$) ns; non-significant ($p>0.05$)

C) Surface roughness test:

Roughness change (Ra) results (Mean±SD) for both denture base materials groups as function of silver (Ag) and Zirconium oxide nanoparticle modification and concentration are summarized in table (3).

With control subgroup; it was found that Flexible group recorded statistically significant higher mean value ($0.2400\ \mu\text{m}$) than Acrylic Resin group ($0.1540\ \mu\text{m}$) as indicated by unpaired t-test ($P=0.005 < 0.05$).

With 2% Ag modified subgroup; it was found that Acrylic Resin group recorded statistically non-significant higher mean value ($0.2160\ \mu\text{m}$)

than Flexible group ($0.2121\ \mu\text{m}$) as indicated by unpaired t-test ($P=0.8863>0.05$).

With 5% Ag modified subgroup; it was found that Acrylic Resin group recorded statistically non-significant higher mean value ($0.2343\ \mu\text{m}$) than Flexible group ($0.2170\ \mu\text{m}$) as indicated by unpaired t-test ($P=0.5949>0.05$).

With 2% Zro modified subgroup; it was found that Flexible group recorded statistically non-significant higher mean value ($0.2193\ \mu\text{m}$) than Acrylic Resin group ($0.1848\ \mu\text{m}$) as indicated by unpaired t-test ($P=0.7442<0.05$).

With 5% Zro modified subgroup; it was found that Flexible group recorded statistically non-significant higher mean value ($0.2242\ \mu\text{m}$) than Acrylic Resin group ($0.1899\ \mu\text{m}$) as indicated by unpaired t-test ($P=0.4201>0.05$).

Total effect of material type

Regardless to nano-particle modification or concentration, totally it was found that Flexible group recorded statistically non-significant higher mean value than Acrylic Resin group as indicated by three-way ANOVA-test ($P=0.0855>0.05$).

Total effect of nano-particle type

Regardless to material group or nano-particle concentration, totally it was found that Ag modified subgroup recorded statistically non-significant higher mean value than Zro modified subgroup as indicated by three-way ANOVA-test ($P=0.5788 >0.05$).

Total effect of nano-particle concentration

Regardless to material group or nano-particle type, totally it was found that 2% modified subgroup recorded statistically non-significant higher mean value than 5% modified subgroup as indicated by three-way ANOVA-test ($P=0.4001 >0.05$).

Table 3: Roughness results (Mean±SD) for both denture base materials groups as function of silver and Zirconium oxide nanoparticles modification and concentration

Control		Acrylic Resin		Flexible		Statistics
		Mean	SD	Mean	SD	P value
		0.1540	0.024	0.2400	0.01	0.005*
Ag modified	2%	0.2160	0.067	0.2121	0.065	0.8863ns
	5%	0.2343	0.073	0.2170	0.072	0.5949ns
ZrO modified	2%	0.1848	0.068	0.2193	0.103	0.7442ns
	5%	0.1899	0.069	0.2242	0.057	0.4201ns
Statistics	P value	0.1573ns		0.8592ns		

*; significant ($p < 0.05$)

ns; non-significant ($p > 0.05$)

DISCUSSION

Flexural strength

Flexural strength is an important property for dental resins, as the major cause of prosthesis fracture in the oral cavity is related to stresses caused by repeated application of masticator forces they induce more bending movements^(40,41). The size shape and distribution of filler particles in the polymer matrix, strong adhesion at the interface and degree of cure play a major role on the mechanical properties of particulate filled polymer composites^(42,43). Korkmaz et al.,⁽⁴⁴⁾ suggested that the size of filler particles should be small for proper processing. The average of particle size of PMMA beads used in commercially available denture base resins is around 100 μm ⁽⁴⁵⁾. The particle size of AgNPs and ZrONPs (20-40 nm) used in this study is much smaller than that of powder resin particles. Thus, AgNPs and ZrONPs will fill the interstitial of polymer particles to give a heterogenous mixture and will not force the displacement of the segments of polymer chain. In addition, low percentage of nanoparticles should be used to ensure that they will be embedded in resin. The degree of the particles dispersion in the matrix is an important factor that influences the strength properties. It is evident that high surface area of the nanoparticles in

PMMA makes the applied stress easily transformed from the matrix onto the nanoparticles, resulting in an enhancement of the mechanical properties. Furthermore, the compatibility between the polymeric matrix and the nanoparticles is improved due to formation of more favourable polar interactions between C=O groups of the PMMA chains and nanoparticles^(46,47), possibly increasing the mechanical strength. Additionally, degree of cure (DC) is the other factor that affects the mechanical properties. Lower DC is usually associated with poorer mechanical properties⁽⁴⁸⁾. More amount of unreacted monomer may act as plasticizer and decreases mechanical properties. Results of this study suggest that addition of AgNPs and ZrONPs alters flexural strength of PMMA depending on the amount of nanoparticles employed. However, the flexural strength of all groups used in this study is more than the ISO 20795-1:2008 (ISO, 2008) requirement. According to ISO 20795-1:2008 (ISO, 2008), the flexural strength of polymeric materials must be at least 50 MPa.

In our present study, with control subgroup; it was found that Acrylic Resin group recorded higher flexural strength than Flexible group. With Ag modified subgroup (2% and 5%); it was found that Acrylic resin group recorded higher value than



Flexible group. Also we found that increasing the AgNPs concentration led to an increase in the flexural strength of both denture base materials than their control groups. This may be due to the degree of nanoparticles dispersion in the PMMA resin matrix and the chemical composition of resin materials used in the study. From this study it can be believed that the polar interactions between the nanoparticles and C=O of polymer matrix (weak interactions) are adequate in improving the flexural strength. This is due to the fact that metal nanoparticles may bond with polymer molecules, crosslinking them between each other and, thereby, increasing the “pseudocrystallinity” of the polymer and forming an ordered system.

These results are in agreement with Abdulkareem MM et al.,⁽⁴⁹⁾ who demonstrated an increase in the flexural strength of microwave cured acrylic denture base material by increasing the concentration of silver nanofillers. Explicitly, it is evident that the chemical composition of acrylic resin, chemical interaction between nanoparticle and C=O group of PMMA polymer, filler particle size and DC will influence the flexural strength of PMMA based acrylic denture base materials.

These results are in disagreement with Muzalev pA. et al.,⁽⁵⁰⁾ who found that all unmodified (control) specimens showed more flexural strength than the modified specimens except Luciton199 and there is a gradual decrease in flexural strength was observed in modified Trevlon specimens from 0.5 wt% to 5.0 wt% AgNPs.

This difference may be due to using different commercial acrylic resin material types, different nanoparticles concentrations or different methods.

With ZrO modified subgroup; it was found that Acrylic Resin group recorded higher value than Flexible group. But in both denture base materials, the flexural strength decrease with increasing ZrONPs concentrations. This may be due to that

increasing in the concentration of nanoparticles in the polymer reduces the chemisorption interaction of the metal nanoparticle with the polymer, which leads to the disordered structure of the nanocomposite and reduces the mechanical strength. Also at higher concentrations, nanoparticles may also act as impurities of polymerization that causes decrease in DC. Hence, more amount of unreacted monomer may be left over within the matrix that results in decreasing the flexural strength.

These results are in agreement with Al-Rais RM, Al-Nakkash WA and Al-Bakri AA-WK.⁽⁵¹⁾ They found that a slight decrease in flexural strength was also reported; it may result from clustering of the particles within the resin, which weakened the material.

These results are in disagreement with Kul E, Aladağ Lİ and Yesildal R.⁽⁵²⁾ They mentioned that incorporating zirconia (ZrO₂) fillers in PMMA significantly increased its flexural strength. This difference may be due to using different commercial acrylic resin material types, different nanoparticles concentrations or different methods.

Color stability test

The change in the appearance and staining indicates reduction of the long term quality of the prosthesis⁽⁵³⁾. The addition of non-transparent fillers to the PMMA for the purpose of improving mechanical properties would induce increase in the amount of light absorption as measured by spectrophotometric device which detect the amount of staining on the prosthesis⁽⁵⁴⁾.

Methods used for measuring color stability of denture base materials includes visual assessment, digital image processing, slide projection, visualization of color spaces and assigning orders, and color comparison^(55,56).

In this study, colors were measured using a Reflective spectrophotometer and measurements

were made according to the CIE Lab color systems for color difference analysis. The CIE Lab color system is comprised of all colors, including light source colors; further, it is not mediated by human perception. This system is recommended for material color difference tests because it is the most scientific color system^(55,57).

The results of this study revealed that significant color shifts occurred in all tested specimens. The increase in light absorption is statistically significant; there was an increase in the relative amount of light absorption with the increasing of ZrO NPS concentration. This is obviously due to the presence of opaque nano-ZrO powder in the polymer matrix which absorbs more light energy than polymer matrix and appears more opaque. These findings were due to the high atomic number of Zr compared to the chemical constituent of acrylic which has low atomic number. The absorption of light energy by an element is dependent chiefly on the cube of its atomic number⁽⁵⁸⁾.

The results of the current study is in agreement with the findings of Ihab N. et al.,⁽⁵⁴⁾ who found that significant color differences were detected between control group and specimens incorporated with zirconium oxide nano-fillers at different immersion solutions.

It is well known that discoloration is a common problem for silver-containing materials because of the oxidative reaction; therefore, further studies are required to investigate the improvement of color stability for the clinical application possibility so as not to compromise the aesthetics of the prosthesis⁽⁵⁹⁾.

The color difference (ΔE) of tested specimens ranged from 7.17 ± 0.44 to 23.19 ± 0.39 compared with control, whereas a color change is considered very small if ΔE is < 1 ⁽⁶⁰⁾; it is clinically acceptable if the color change is between 1 and 2. It is considered clinically perceptible if the ΔE is > 3.3 ⁽⁶¹⁾ and indicates a poor match and clinically unacceptable if the ΔE is > 3.7 ^(62,63). It was observed that significantly color difference (ΔE) existed between specimens in

each tested group that was all perceivable by human eye and that difference was clinically unacceptable (ΔE is > 3.7).

Surface roughness test

We evaluated the effect of Ag and ZrO₂ nanoparticles addition on the surface roughness of the acrylic resin material. The surface roughness of denture material is important, because it affects the oral health of tissues in direct contact with the dentures^(20,21). The surface roughness threshold for acrylic resin is $0.2 \mu\text{m}$, below which no significant decrease in bacterial colonization occurs⁽⁶⁴⁾. Dramatic colonization would be expected to occur on surfaces with a roughness value of $2.2 \mu\text{m}$ ⁽⁶⁵⁾. The surface roughness of polished acrylic resin varies between $0.03 \mu\text{m}$ and $0.75 \mu\text{m}$. However, an important factor in the clinical performance of a material is the way it responds to hygiene procedures⁽⁶⁶⁾. In agreement with the study of Saad-Eldeen et al.,⁽¹⁸⁾ the results of our study showed that incorporating Ag and ZrO₂ nanoparticles at different concentrations did not adversely affect the roughness of the denture base resin.

The slight changes of the surface roughness of the modified specimens may be due to the presence of the Ag and ZrO₂ nanoparticles at the surface which acts as fillers among the interpolymeric chains and fills the voids in the body of the specimens. Also, this may be due to of finishing and polishing of the specimens. The changes of the surface roughness agreed with Ihab N.⁽³⁴⁾ since they found that the addition of nano-ZrO₂ fillers into the acrylic denture base did not significantly change the surface roughness when different percentages of modified acrylic denture base material. But the results of the current study is in disagreement with the findings of Vojdani et al.,⁽¹⁵⁾ who concluded that incorporation of micron-sized Al₂O₃ powder in the heat cured acrylic resin produced a slight increase in the surface roughness. This may be due to the fillers used which are different from the fillers in the present study.



CONCLUSION

- 1- The effect of nanoparticles on flexural strength depends on several factors including the type of denture base material and also the type and the concentrations of nanoparticles used.
- 2- Significant color differences were detected between control group and specimens incorporated with (Ag & ZrO) nanoparticles at different concentration and these differences were clinically unacceptable.
- 3- Surface roughness of all tested groups show non-significant statistical differences when using different nanoparticles type or concentration.

REFERENCES

1. Zarb GA, Hobkirk J, Eckert S, Jacob R. Prosthodontic treatment for edentulous patients: complete dentures and implant-supported prostheses: Elsevier Health Sciences; 2013.
2. Vojdani M, Khaledi A. Transverse strength of reinforced denture base resin with metal wire and E-glass fibers. Journal of Dentistry of Tehran University of Medical Sciences. 2006;3(4):159-66.
3. Lowe LG. Flexible denture flanges for patients exhibiting undercut tuberosities and reduced width of the buccal vestibule: a clinical report. The Journal of prosthetic dentistry. 2004; 92(2):128-31.
4. Phoenix RD, Mansueto MA, Ackerman NA, Jones RE. Evaluation of mechanical and thermal properties of commonly used denture base resins. Journal of Prosthodontics. 2004;13(1):17-27.
5. Negrutiu M, Sinescu C, Romanu M, Pop D, Lakatos S. Thermoplastic resins for flexible framework removable partial dentures. TMJ. 2005;55(3):295-9.
6. Al-Noori AK, Hatim NA, Taqa AA. Thermal diffusivity of nano-sized additives on flexible denture base material. Int J Enh Res Sci Tech & Eng. 2015;4(4):210-7.
7. Vallittu P, Lassila V. Effect of metal strengthener's surface roughness on fracture resistance of acrylic denture base material. Journal of oral rehabilitation. 1992;19(4):385-91.
8. Vallittu P. Effect of some properties of metal strengtheners on the fracture resistance of acrylic denture base material construction. Journal of oral rehabilitation. 1993;20(3):241-8.
9. Robinson McCabe J. Impact strength of acrylic resin denture base materials with surface defects. Dent Mater. 1993;9:355-60.
10. Matsukawa S, Hayakawa T, Nemoto K. Development of high-toughness resin for dental applications. Dental Materials. 1994;10(6):343-6.
11. Kim S-H, Watts DC. The effect of reinforcement with woven E-glass fibers on the impact strength of complete dentures fabricated with high-impact acrylic resin. The journal of prosthetic dentistry. 2004;91(3):274-80.
12. Uzun G, Hersek N, Tincer T. Effect of five woven fiber reinforcements on the impact and transverse strength of a denture base resin. The Journal of prosthetic dentistry. 1999;81(5):616-20.
13. Chen SY, Liang WM, Yen PS. Reinforcement of acrylic denture base resin by incorporation of various fibers. Journal of Biomedical Materials Research: An Official Journal of The Society for Biomaterials, The Japanese Society for Biomaterials, and The Australian Society for Biomaterials and the Korean Society for Biomaterials. 2001;58(2):203-8.
14. Jagger D, Harrison A, Jandt K. The reinforcement of dentures. Journal of oral rehabilitation. 1999;26(3):185-94.
15. Vojdani M, Bagheri R, Khaledi AAR. Effects of aluminum oxide addition on the flexural strength, surface hardness, and roughness of heat-polymerized acrylic resin. Journal of dental sciences. 2012;7(3):238-44.
16. Ayad NM, Badawi MF, Fatah AA. Effect of reinforcement of high-impact acrylic resin with zirconia on some physical and mechanical properties. Archives of Oral Research. 2008;4(3).
17. Panyayong W, Oshida Y, Andres C, Barco T, Brown D, Hovijitra S. Reinforcement of acrylic resins for provisional fixed restorations. Part III: effects of addition of titania and zirconia mixtures on some mechanical and physical properties. Bio-medical materials and engineering. 2002;12(4):353-66.
18. Saad-Eldeen MA, AL-Fallal A, Abouelatta O. Effect of zirconium oxide reinforcement on epithelial oral mucosa, Immunoglobulin and surface roughness of complete acrylic heat-cured denture. Egypt Dent Associat. 2007;53:941-6.
19. Abdel-Samad A, EL-Fallal A. Evaluation of the effect of zirconium oxide on wear resistance and hardness of acrylic teeth. Egypt Dent J. 2009;55:639-43.

20. Radford DR, Sweet S, Challacombe S, Walter J. Adherence of *Candida albicans* to denture-base materials with different surface finishes. *Journal of dentistry*. 1998;26(7):577-83.
21. Verran J, Maryan CJ. Retention of *Candida albicans* on acrylic resin and silicone of different surface topography. *The Journal of prosthetic dentistry*. 1997;77(5):535-9.
22. Jordan J, Jacob KI, Tannenbaum R, Sharaf MA, Jasiuk I. Experimental trends in polymer nanocomposites—a review. *Materials science and engineering: A*. 2005;393(1-2):1-11.
23. Molefi J, Luyt A, Krupa I. Comparison of the influence of Cu micro-and nano-particles on the thermal properties of polyethylene/Cu composites. *eXPRESS Polymer Letters*. 2009;3(10):639-49.
24. Sondi I, Salopek-Sondi B. Silver nanoparticles as antimicrobial agent: a case study on *E. coli* as a model for Gram-negative bacteria. *Journal of colloid and interface science*. 2004;275(1):177-82.
25. Katakam S, Krishna DSR, Kumar TS. Microwave processing of functionally graded bioactive materials. *Materials Letters*. 2003;57(18):2716-21.
26. Shi G-M, Han J-K, Zhang Z-D, Song H-Y, Lee B-T. Pretreatment effect on the synthesis of Ag-coated Al₂O₃ powders by electroless deposition process. *Surface and Coatings Technology*. 2005;195(2-3):333-7.
27. Alt V, Bechert T, Steinrücke P, Wagener M, Seidel P, Dingeldein E, et al. An in vitro assessment of the antibacterial properties and cytotoxicity of nanoparticulate silver bone cement. *Biomaterials*. 2004;25(18):4383-91.
28. She W. Basic study of denture base resin with nano-silver antibacterial agent. *Dent Mater J*. 2004;27:176-80.
29. Chen L, Huang Z, Xue C. PC nanofiber reinforced PMMA transparent composites incorporated with TiO₂ nanoparticles. *J Inorg Mater*. 2009;24:469-74.
30. Hoskins JS, Karanfil T, Serkiz SM. Removal and sequestration of iodide using silver-impregnated activated carbon. *Environmental science & technology*. 2002; 36(4):784-9.
31. Damm C, Münstedt H, Rösch A. Long-term antimicrobial polyamide 6/silver-nanocomposites. *Journal of Materials Science*. 2007;42(15):6067-73.
32. Damm C. Silver ion release from polymethyl methacrylate silver nanocomposites. *Polymers and Polymer Composites*. 2005;13(7):649-56.
33. Mitra SB, Wu D, Holmes BN. An application of nanotechnology in advanced dental materials. *The Journal of the American Dental Association*. 2003;134(10):1382-90.
34. Ihab N. Evaluation the effect of modified nano-fillers addition on some properties of heat cured acrylic denture base material. *Journal of Baghdad college of dentistry*. 2011;23(3):23-9.
35. Gad MM, Rahoma A, Al-Thobity AM, ArRejaie AS. Influence of incorporation of ZrO₂ nanoparticles on the repair strength of polymethyl methacrylate denture bases. *International journal of nanomedicine*. 2016;11:5633.
36. Xin-jing Z, Xiu-yin Z, Bang-shang Z, Lu Z, Chen Q. Effect of nano ZrO₂ on flexural strength and surface hardness of polymethylmethacrylate. *Shanghai Journal of Stomatology*. 2011;20(4).
37. Gad M, ArRejaie AS, Abdel-Halim MS, Rahoma A. The reinforcement effect of nano-zirconia on the transverse strength of repaired acrylic denture base. *International journal of dentistry*. 2016;2016.
38. Salih SI, Oleiwi JK, Hamad QA. Investigation of fatigue and compression strength for the PMMA reinforced by different system for denture applications. *International Journal of Biomedical Materials Research*. 2015;3(1):5-13.
39. Ahmed MA, Ebrahim MI. 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*. 2014;4(02):50.
40. Alla RK, Sajjan S, Alluri VR, Ginjupalli K, Upadhya N. Influence of fiber reinforcement on the properties of denture base resins. *Journal of Biomaterials and Nanobiotechnology*. 2013;4(1):91.
41. Shakeel S, Alla RK, Shamma M, Ahmed T. Evaluation and comparison of the transverse strength of two heat cure denture base resins repaired with auto polymerizing resin by wetting with Methyl Methacrylate (MMA) at different time intervals—An in vitro study. *British Journal of Medicine and Medical Research*. 2015;7(4):263.
42. Garoushi SK, Lassila L, Vallittu PK. Short fiber reinforced composite: the effect of fiber length and volume fraction. *J Contemp Dent Pract*. 2006;7(5):10-7.
43. Unal H, Mimaroglu A. Influence of filler addition on the mechanical properties of nylon-6 polymer. *Journal of reinforced plastics and composites*. 2004;23(5):461-9.
44. Korkmaz T, Doğan A, Usanmaz A. Dynamic mechanical analysis of provisional resin materials reinforced by metal oxides. *Bio-medical materials and engineering*. 2005;15(3):179-88.



45. Alla RK, Swamy KR, Vyas R, Konakanchi A, Guduri V, Gadde P. Influence of Silver Nanoparticles Incorporation on Flexural Strength of Heat-cure Acrylic Denture Base Resin Materials. *ANNUAL RESEARCH & REVIEW IN BIOLOGY*. 2017;17(4).
46. Makvandi P, Nikfarjam N, Sanjani NS, Qazvini NT. Effect of silver nanoparticle on the properties of poly (methyl methacrylate) nanocomposite network made by in situ photoiniferter-mediated photopolymerization. *Bulletin of Materials Science*. 2015;38(6):1625-31.
47. Kassaee M, Mohammadkhani M, Akhavan A, Mohammadi R. In situ formation of silver nanoparticles in PMMA via reduction of silver ions by butylated hydroxytoluene. *Structural Chemistry*. 2011;22(1):11-5.
48. Zhang Y, Chen Y-y, Huang L, Chai Z-g, Shen L-j, Xiao Y-h. The antifungal effects and mechanical properties of silver bromide/cationic polymer nano-composite-modified Poly-methyl methacrylate-based dental resin. *Scientific Reports*. 2017;7(1):1547.
49. Abdulkareem MM, Hatim NA. The effect of adding metallic nano fillers on flexural strength of heat cure acrylic resin treated by microwave. *International Journal of Enhanced Research in Science, Technology & Engineering*. 2016;5(8):17-25.
50. Muzalev P, Kosobudskii I, Ushakov N, Panova L. Metal nanoparticles in acrylic polymer matrices. *Inorganic Materials: Applied Research*. 2011;2(5):528-30.
51. Al-Rais RM, Al-Nakkash WA, Al-Bakri AA-WK. Filler reinforced acrylic denture base material. Part 2-Effect of water sorption on dimensional changes and transverse strength. *Journal of baghdad college of dentistry*. 2005;17(1):6-10.
52. Kul E, Aladağ Lİ, Yesildal R. Evaluation of thermal conductivity and flexural strength properties of poly (methyl methacrylate) denture base material reinforced with different fillers. *The Journal of prosthetic dentistry*. 2016;116(5):803-10.
53. Szabó G, Valderhaug J, Ruyter IE. Some properties of a denture acrylic coating. *Acta Odontologica Scandinavica*. 1985;43(4):249-56.
54. Ihab N, Hassanen K, Ali N. Assessment of zirconium oxide nano-fillers incorporation and silanation on impact, tensile strength and color alteration of heat polymerized acrylic resin. *Journal of baghdad college of dentistry*. 2012;24(special issue 2):36-42.
55. Sepúlveda-Navarro WF, Arana-Correa BE, Ferreira Borges CP, Habib Jorge J, Urban VM, Campanha NH. Color stability of resins and nylon as denture base material in beverages. *Journal of Prosthodontics: Implant, Esthetic and Reconstructive Dentistry*. 2011;20(8):632-8.
56. Paul S, Peter A, Pietrobon N, Hämmerle C. Visual and spectrophotometric shade analysis of human teeth. *Journal of dental research*. 2002;81(8):578-82.
57. Hersek N, Uzun G, Yildiz P. Color stability of denture base acrylic resins in three food colorants. *Journal of Prosthetic Dentistry*. 1999;81(4):375-9.
58. Stafford G, MacCulloch W. Radiopaque denture base materials. *British dental journal*. 1971;131(1):22.
59. Nam KY, Lee CH, Lee CJ. Antifungal and physical characteristics of modified denture base acrylic incorporated with silver nanoparticles. *Gerodontology*. 2012;29(2):e413-e9.
60. O'brien W, Groh C, Boenke K. A new, small-color-difference equation for dental shades. *Journal of dental research*. 1990;69(11):1762-4.
61. Aziz T, Waters M, Jagger R. Development of a new poly (dimethylsiloxane) maxillofacial prosthetic material. *Journal of Biomedical Materials Research Part B: Applied Biomaterials: An Official Journal of The Society for Biomaterials, The Japanese Society for Biomaterials, and The Australian Society for Biomaterials and the Korean Society for Biomaterials*. 2003;65(2):252-61.
62. Johnston W, Kao E. Assessment of appearance match by visual observation and clinical colorimetry. *Journal of dental research*. 1989;68(5):819-22.
63. Goldstein GR, Schmitt GW. Repeatability of a specially designed intraoral colorimeter. *The Journal of prosthetic dentistry*. 1993;69(6):616-9.
64. Bollenl CM, Lambrechts P, Quirynen M. Comparison of surface roughness of oral hard materials to the threshold surface roughness for bacterial plaque retention: a review of the literature. *Dental materials*. 1997;13(4):258-69.
65. Quirynen M, Marechal M, Busscher H, Weerkamp A, Darius P, van Steenberghe D. The influence of surface free energy and surface roughness on early plaque formation: an in vivo study in man. *Journal of clinical periodontology*. 1990;17(3):138-44.
66. Busscher H, Van Pelt A, De Boer P, De Jong H, Arends J. The effect of surface roughening of polymers on measured contact angles of liquids. *Colloids and Surfaces*. 1984;9(4):319-31.