

MICROTENSILE BOND STRENGTH OF ZIRCONIA NANOFILLERS REINFORCED ACRYLIC RESIN TO DENTURE TEETH AFTER THERMOCYCLING

Kawkb Mohamed Eltamimi * and Dalia Abdel-Moniem Bayoumi **

ABSTRACT

Statement of problem: Addition of zirconium oxide nanofillers to PMMA had improved the mechanical properties of heat polymerized acrylic resin, but its effect on bond strength to acrylic teeth hadn't been evaluated yet.

Purpose: The purpose of the study was to evaluate the effect of Zirconium oxide (ZrO_2) nanofillers powder with different concentrations (1.5%, 3%, 5%) on bond strength of resin denture base to acrylic teeth before and after thermocycling.

Material and methods: Zirconium oxide nanoparticles powder were added to heat cured (PMMA) with different concentrations (0, 1.5%, 3%, 5%). The base surfaces of 16 molar acrylic teeth were flattened, and zirconia reinforced heat-polymerized acrylic resin was applied to them. Thereafter, bar specimens were produced for the microtensile bond strength testing either before or after thermocycling.

Results: The results showed that 5% ZrO_2 conc. recorded the highest value of microtensile bond strength. The lowest value was recorded in 1.5%, followed by 3%. Regarding thermocycling, the non-thermocycled groups always recorded higher values of microtensile bond strength either significant (1.5% & 5%) or insignificant (0% & 3%) than thermocycled ones at $p < 0.05$.

Conclusions: Within the limitation of this study, it was concluded that reinforcement of acrylic denture base with zirconia nanofillers had significantly decreased bond strength with acrylic denture teeth in 1.5 % and 3% concentration; however, 5% had increased the bond strength values insignificantly. Thermocycling had reduced the bond strength in all concentrations.

KEYWORDS: Zirconium oxide nanofillers, microtensile bond strength, thermocycling, acrylic resin and acrylic denture teeth.

* Lecturer of Prosthodontics, Faculty of Dentistry, Suez Canal University

** Lecturer of Dental Materials, Faculty of Dentistry, Suez Canal University

INTRODUCTION

Acrylic resin polymethyl methacrylate (PMMA) has been the most prevalent material for the fabrication of dentures. It has many advantages such as good aesthetics, precise fit, steadiness in the oral environment, simple processing technique and low cost fabrication.⁽¹⁾ However, this material is still deficient to achieve the ideal mechanical requirements for dental applications, mainly due to its low fracture resistance and plaque accumulation.^(2,3)

Different fillers, metal oxides, and carbon graphite fibers are blended into the composition to improve the mechanical properties of the resin.^(4,5) One of the most promising newly used fillers to overcome strength deficiency is the addition of nanoparticles into PMMA to act as the reinforcing material.⁽⁶⁾

Numerous nanoparticles has been used such as aluminum oxide (Al_2O_3), zirconium oxide (ZrO_2), titanium dioxide (TiO_2), zinc oxide (ZnO), silicon dioxide (SiO_2) and silver (Ag).^(7,8) Nanoparticles are advantageous in their small size, large surface area, and intense interface interactions with the polymer matrix. Thus, nanoparticles improve the physical and optical properties of the polymer matrix. In addition, they can resist the environmental stress, cracking and aging.⁽⁸⁾

Zirconium oxide nanoparticles (ZrO_2 NPs) have excellent biocompatibility and superior aesthetics than other metal oxide ones.⁽⁴⁾

Previous studies have reported that 20 to 33% of dentures' repair is related to artificial teeth detaching from denture bases or teeth breaking off.⁽⁹⁻¹²⁾ The bonding mechanism between acrylic resin denture bases and artificial teeth is dependent on a polymer interpenetrating network or chemical covalent bond.⁽¹³⁾

The effect of zirconium oxide nanoparticles to PMMA denture bases was not thoroughly investigated. Therefore, the purpose of this study was to investigate the effect of addition of these nanoparticles powder in different percentages to evaluate the bond strength of acrylic teeth to resin denture base.

MATERIALS AND METHODS

Nano ZrO_2 powder particles of 99.9% purity (Nanogate, Egypt) and an average granularity of 90 nm and surface area of $12 \pm 3 \text{ m}^2/\text{g}$. was added to heat polymerized PMMA powder (Acrostone, Anglo-Egyptian Company, Hegaz, Cairo, Egypt) in various concentration (1.5%, 3% and 5%) .

Salinization of nano- ZrO_2 particles

Nano ZrO_2 particles were treated by silane coupling agent TMSPM (3-trimethoxysilyl) propyl methacrylate (SIGMA-ALDRICH, Germany). A 0.3 g of TMSPM was dissolved in 100 mL of acetone to grantee and 30 grams of nano- ZrO_2 particles were added to the TMSPM/acetone solution and stirred with a magnetic stirrer (Hot plate with magnetic stirrer, MSH-A 30A, South Korea) for 60min to evenly coat the nanoparticles surfaces. Then, a rotary evaporator was used to remove the solvent under vacuum at 60°C and 150 rpm for 30 min. The dried powder was heated at 120°C for 2 h and then left to cool to room temperature.^{(14,15)*}

PMMA/ ZrO_2 nanocomposite preparation

By using an electronic balance (Scaletec instruments heiligenstadt German), the silanized ZrO_2 nanoparticles were added to PMMA powder in concentrations 1.5%, 3% and 5% by weight. Every pre-weighed nano-filler powder in the determined concentration were added individually to PMMA powder and thoroughly mixed using a mortar and

* Salinization process was carried out in lab of Inorganic Chemistry, Chemistry Department, Faculty of Science, Suez Canal University by Prof. Dr. Sabry A. El-Korashy

pestle and stirred for 30 min to achieve an equal distribution of nanoparticles. ^(16,17)

The base surfaces of 16 acrylic teeth were flatted in a polishing machine using silicon carbide papers with grits of 600, 800, and 1,000 under cooling. The artificial teeth were fixed to a metallic base with wax while the surfaces to be flattened facing down and surrounded by polyvinyl chloride (PVC) rings of 18 mm height and 17 mm diameter that were fixed to the metallic base using cyanoacrylate adhesive. Then, dental plaster (Dental plaster, De Juang, China) was mixed following the manufacturer's instructions and poured inside the rings under vibration. After setting, the assemblies were separated from the metallic base. The PVC rings were then cut longitudinally with a carborundum disk and removed. The plaster blocks containing the embedded teeth carefully removed from the tubes and placed on a glass slab with the prepared flat surfaces facing up. A second PVC ring was positioned over the plaster block, encircling the tooth sample, and filled with molten wax to create mold to be filled by the denture base resin. The plaster-tooth-wax assemblies were placed in a flask base with the wax turned upward. Dental stone (Zeta Mufle, Nevilicure, Italy) was poured up to the plaster-wax border. The stone was set, and the remaining part of the flask was poured with plaster. The flask was then pressed (1,250 kgf) for 45 minutes. After setting, the flask was placed in boiling water for 10 minutes. Afterwards, the wax was eliminated by thoroughly washing it with boiling water and anionic detergent (Pril, Henkel, Egypt). The heat-polymerized acrylic resin was mixed according to manufacturer's recommendations and applied on the denture teeth. The flasks were pressed in a hydraulic press (1,250 kgf) for 8 hours, and resin was polymerized at 74°C for 9 hours. After heat curing, the flasks were kept cooling till room temperature.

Finally, the specimens were removed from the flask and stored in distilled water at 37°C for 7 days.

Thermocycling

Half of the samples of each group were thermocycled in thermocycler (1100 SD Mechatronics thermocycler, Germany) in range of 5°C- 55°C with dwell time 60s. up to 3000 cycles. This procedure corresponded to a 3-year period of oral temperature conditions. ^(18,19)

Production of Beam Specimens

Each tooth-acrylic resin set was serially sectioned to obtain rectangular specimens with mean cross-sectional area of 1 mm². The specimens were fixed to a metal base that was coupled to a cutting machine and then sectioned under water cooling parallel to the long axis of the tooth and perpendicular to the bonding interface using a slow-speed diamond disk in a cutting machine (Isomet 4000, Buehler Ltd., Lake Bluff, IL, USA). The peripheral slices (0.5mm) were discarded because of irregularities at the interface. Twenty-five untrimmed bar specimens 1 mm in thickness and 5.5 mm in length were obtained for each group, (Fig. 1).

Micro tensile Bond Strength Test

The dimensions of the specimens were measured with a digital caliper ((Mitutoyo, Tokyo, Japan) to ensure bonded area of 1 mm². Each specimen beam was aligned in the central groove of a jig and glued in place by its ends using cyanoacrylate-based glue (Zapit, DVA Inc, USA). The tensile test was performed using testing machine (Instron, MA, USA). The calculated bond strength (MPa) = L/A, where L = load (N) for detaching of the specimen and A = interfacial area surface (1mm²).

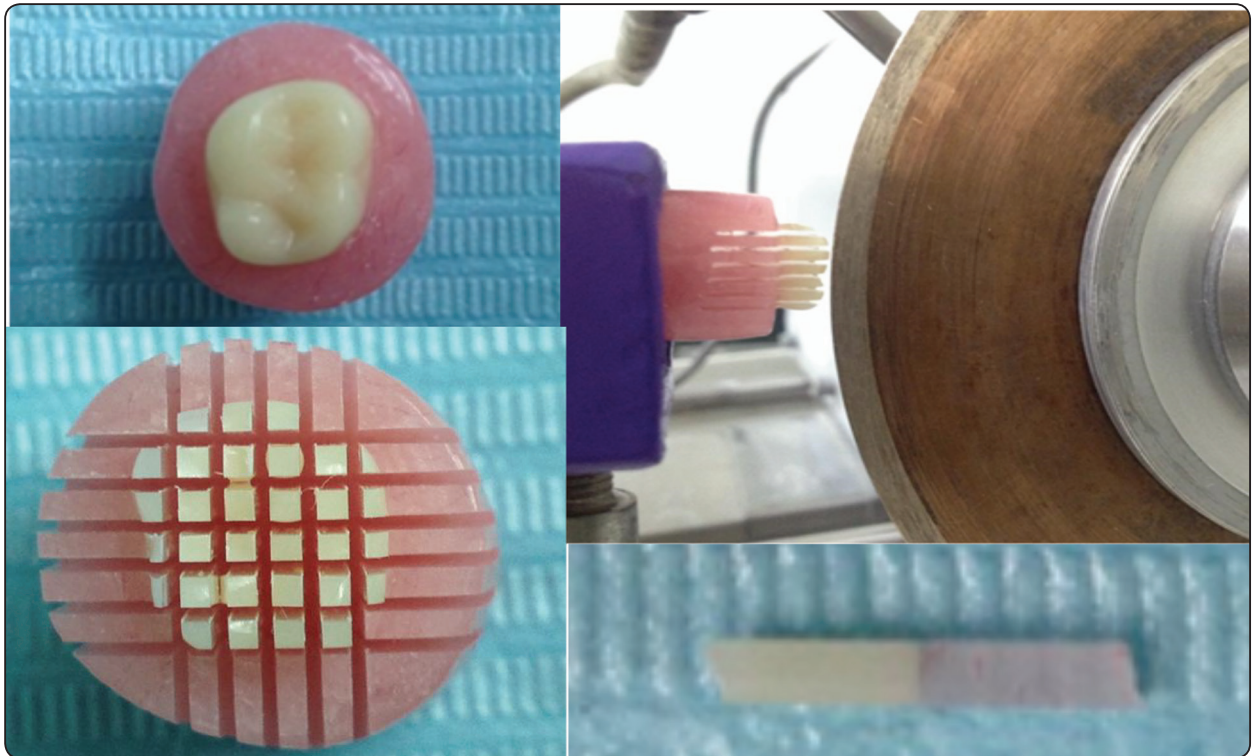


Fig. (1) (a) Tooth-resin assembly; (b) diamond disk; (c) beam specimens(d) bar specimens.

Statistical analysis

The mean and standard deviation of Microtensile stress at Maximum Load (MPa) were calculated. The Kolmogorov-Smirnov, Shapiro-Wilk, Independent T-test and two ways-ANOVA tests were performed using SPSS 23. The Kolmogorov-Smirnov and Shapiro-Wilk tests were performed to check the normality of the data. The two ways-ANOVA tests was carried out to identify significant variance of the studied variable at $p < 0.05$ was used. In addition, the Levene's test with the significant level at $p < 0.05$ was adopted to determine the homogeneity of variances in terms of ANOVA tests. Independent T-test was performed for comparison of the mean differences between the two subgroups thermocycled and nonthermocycled at the same concentration as the cut-off for significance (confidence level in

95%). The bond strength data were analyzed using 2-way analysis of variance (ANOVA) ($P = .05$) and Tukey post hoc test.

RESULTS

Table (1) analysis of variance for microtensile bond strength showed significant difference between thermocycled and non thermocycled, concentrations of zirconium oxide and their interactions at $p < 0.05$.

According to concentration of zirconium oxide, 5% conc. recorded the highest value of microtensile bond strength (38.31 MPa) followed by zero% concentration (36.62 MPa). On the other hand, the lowest value was recorded in 1.5 % (27.37 MPa) followed by 3% concentration with value (28.89 MPa), Table (2) and Figure (2).

TABLE (1) Two-Way ANOVA of Microtensile Bond Strength Data

Source of variance	D.F	S.S	M.S	F	P value	Sig
Thermocycled and nonthermocycled	1	130.77	130.77	6.56	0.015	*
Concentration of zirconium oxid	3	1723.02	574.34	28.81	0.000	***
Concentration * thermocycled and nonthermocycled.	3	629.95	209.98	10.53	0.002	**
Experimental Error	192	3827.00	19.93			
Total	199					

d.f. Degrees of freedom *SS* = Sum of square. *MS* = Mean of square *F* = F calculated.

TABLE (2) Mean of the microtensile bond strength results (MPa) before and after thermocycling

Groups	Non thermocycled	Thermocycled	Mean
CON-0	39.88±4.49 ^{ab}	33.36±10.55 ^{bc}	36.62±4.61 ^A
CON-1.5	30.00±5.95 ^{cd}	24.73±7.00 ^d	27.37±3.72 ^B
CON-3	29.15±5.61 ^{cd}	28.62±8.22 ^{cd}	28.89±0.37 ^B
CON-5	42.48±10.27 ^a	34.13±4.09 ^{bc}	38.31±5.90 ^A
Mean	35.38±6.79 ^a	30.21±4.39 ^b	

Different letters means significant difference between non thermocycled and thermocycled in all concentration at P value<0.5

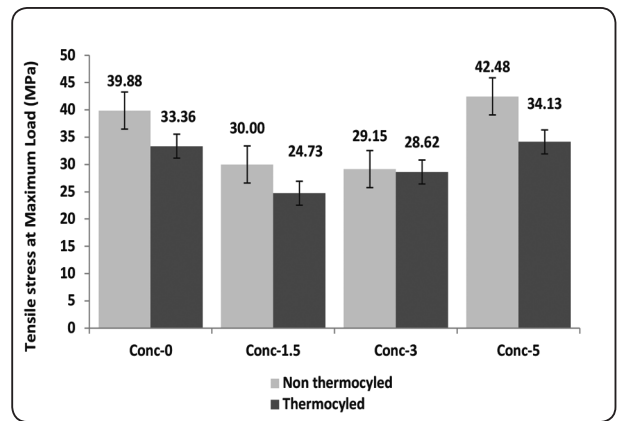


Fig. (2) Means and standard deviation of the microtensile bond strength (MPa) of different Zirconium oxide concentration% before and after thermocycling

TABLE (3) Independent T test between thermocycled and non thermocycled groups

Conc. of ZrO ₂ %	Groups	Mean	± S. D	Mean Difference	95% Confidence Interval		Indep-T-test	Sig. at P value <0.05
					Lower	Upper		
0%	Nonthermocycled	39.88 ^{ab}	4.49	6.52	-2.36	15.41	1.54	0.140
	Thermocycled	33.36 ^{bc}	10.55					
1.5%	Nonthermocycled	30.00 ^{cd}	5.95	5.97	-10.75	-1.18	2.63	0.020**
	Thermocycled	24.73 ^d	7.00					
3%	Nonthermocycled	29.15 ^{cd}	5.61	0.53	-7.50	8.41	0.141	0.890
	Thermocycled	28.62 ^{cd}	8.22					
5%	Nonthermocycled	42.48 ^a	10.27	8.34	-13.45	-3.22	3.60	0.004**
	Thermocycled	34.13 ^{bc}	4.09					

Different letters means significant difference between nonthermocycled and Thermocycled in all concentration at P value<0.5
****; means significant difference between thermocycled and nonthermocycled groups at the same concentration at P value<0.5

Regarding thermocycling, the non-thermocycled groups always recorded higher values of microtensile bond strength either significant (1.5% & 5%) or insignificant (0% & 3%) than thermocycled ones at $p < 0.05$.

DISCUSSION

Acrylic resin PMMA is the popular material for denture base construction since 1937. However, its mechanical properties does not fulfil the required criteria.⁽³⁾ Zirconia nano particles have good toughness and mechanical strength properties.^(16,20,21) It has proven that the addition of zirconium oxide nanoparticles to PMMA increased the flexural strength, fracture toughness, and hardness of heat polymerized acrylic resins as the stresses transfer from weak PMMA matrix to strong nanoparticles.⁽²¹⁾

Though acrylic denture teeth have the advantage of chemical bonding to acrylic denture base, debonding of the teeth away from the dentures is a frequent problem.⁽⁹⁾ Therefore, this study was designed to evaluate the effect of zirconium oxide (ZrO_2) nanofillers powder added in different concentrations (1.5%, 3% and 5%) on bond strength of acrylic teeth to resin denture base.

Chemical bonding depends on the polymerization reaction of the polymeric denture base at tooth-denture base interface. The monomer from denture base diffuses and penetrates into the resin teeth in contact to form an interwoven polymeric network.⁽¹⁰⁾

Despite the well-known of reinforcing effect of ZrO_2 nanoparticles of the strength properties of acrylic denture bases, the result of this study revealed that addition of nano zirconia particles in 1.5% and 3% concentrations significantly decreased bond strength of acrylic resin to resin teeth. This may be referred to the interference of the ZrO_2 nanoparticles with proper adhesion of acrylic resin denture base. The presence of zirconia nanoparticles may impede the diffusion of free monomer in the dough stage to wet, penetrate and swell the base of artificial teeth that hinder proper bonding. i.e. disturb the integrity of the formed polymeric network.⁽¹⁰⁾

The poor dispersion of fillers might be another explanation interpret for the reduction of bond strength. Agglomerated nanoparticles could initiate cracks that propagate up to failure at lower bond strength values than the expected ones. They act as sites of stress raisers, leading to stress concentration.⁽²²⁾ On application of tensile force to the specimens, the stress level at the interface region is considerably higher than the average value providing an easy path for catastrophic failure.⁽²³⁾

On the other hand, the addition of 5% zirconia insignificantly increased the bond strength values when compared with group contained no ZrO_2 (control group). This result is owing to the effect of silane coupling agent. Principally, the silane coupling agent is applied to the zirconia particles to improve the their compatibility with the polymer matrix.^(24,25) Silanes can bond organic material to inorganic one since they have two different functional groups in their chemical structure; an organofunctional group that reacts with an organic resin and an alkoxy groups reacts with the inorganic material permitting adequate adhesion between nanoparticles and the resin matrix.⁽²⁶⁾ This action increased the chemical bond of the interfacial polymer network.⁽²⁷⁾ Consequently, improved the bond strength of teeth with denture base and surpass the debonding forces at the interface. This was confirmed by the dominant cohesive failure of this group.

Thermocycling was used to evaluate the durability of bond and to closely simulate the oral conditions.⁽²⁸⁾ The present results that denoted the deteriorating effect of thermocycling may be attributed to the plasticizing effect of absorbed water that weakened the bond strength as diffusion of water molecules took place through the acrylic tooth-denture base interface. Moreover, hydraulic degradation of nanoparticles due to hydrophilicity of the silane coupling would impair the integrity of the polymeric network. Besides, the thermal stresses during thermocycling augmented the weakening effect on the bond strength.⁽²⁹⁾

CONCLUSIONS

Within the limitation of this study, it was concluded that reinforcement of acrylic denture base with zirconia nanofillers significantly decreased bond strength with acrylic denture teeth in 1.5 % and 3% concentration; however, 5% had increased the bond strength values insignificantly. Thermocycling reduced the bond strength in all concentrations.

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