

## EFFECT OF AGING AND NANO SILICA SURFACE TREATMENT OF ZIRCONIA CERAMICS CROWNS ON MICROLEAKAGE

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### ABSTRACT

**Objective** The goal of this study was to find out how aging and a surface treatment called sol-gel nano silica affect microleakage in zirconia ceramic crowns.

**Materials and methods:** Twenty recently extracted human maxillary first molars were prepared for full coverage zirconia crowns made of Yttrium-stabilized zirconia ceramics that are readily available. They were split into four groups, each with five samples the control group, the thermocycling group, the nanosilica group, and the nanosilica-thermocycling group. Zirconia crown fitting surfaces were coated with a sol-gel nano silica substance. Thermal cycling was performed 5000 thermal cycles. TheraCem resin cement was used to cement all of the crowns. Crowns were submerged in 0.5% methylene blue solution for 24 hours before being sectioned bucco-lingually for microleakage testing. The depth of dye penetration was evaluated using a light stereomicroscope (Nikon MA100 Japan) at 50 magnification and image analysis software (Omnimet, Buchler, USA). The Tukey test and one-way ANOVA were used for statistical analysis.

**Result:** The nanosilica group had the lowest microleakage, followed by the nano-silica with thermocycling group. Among the groups, the thermocycling group had the highest microleakage values.

**Conclusion:** Although sol-gel nano silica coating reduces microleakage, ageing has a negative impact on it.

**KEYWORDS:** microleakage, zirconia ceramics, nano silica, and aging.

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## INTRODUCTION

Ceramics are often used in dentistry because there is a high demand for non-metallic materials that look good and have good qualities like biocompatibility, high bond strength, high wear resistance, and low thermal conductivity.

The most common dental ceramic systems are made of silica, leucite, lithium disilicate, alumina, and zirconia. Zirconia (zirconium dioxide,  $ZrO_2$ ) possesses ideal qualities for dental applications, including exceptional toughness, strength, and fatigue resistance, as well as outstanding wear attributes and biocompatibility<sup>(1)</sup>.

Computer-aided design/computer-aided machining advancements have facilitated the production of zirconia prosthesis. However, because of the glass-free component structure that characterises zirconia as an acid-resistant material, achieving a permanent bond strength between zirconia-based materials and resin cement has proven difficult.<sup>(1)</sup>

Mechanical and/or chemical bonding of resin cement to the zirconia surface can produce a strong, long-lasting bond. Several studies demonstrated that nano silica functioned well in a variety of dental applications. Many silica coating techniques have been developed, including the conventional silica-coated approach, the tribochemically silica-coated methodology, the tentative plasma spray technique, and the vapor-phase deposition technique.<sup>(2)</sup> Even though these processes often require the use of special tools, they result in better zirconia bonds.

Traditional silica coating processes have been criticised for their negative impacts on mechanical characteristics (i.e., causing surface defects and phase transitions), as well as their high cost. As a result, various technologies, such as the sol-gel process, have been used for zirconia ceramic silica coating. A sol-gel approach allows for the direct processing of glasses and thin films from solution without the need of powder. An oxide network arises

during this process as a result of the hydrolysis and condensation of metal alkoxide precursors. Lately the sol-gel technique has been applied for zirconia silica coating to increase resin-zirconia bonding.<sup>(3)</sup>

Many investigations on chemical bonding found that primers and resin cement based on 10-methacryloyloxydecyl dihydrogen phosphate (MDP) monomer enhanced binding strength. They are preferred cementation materials for indirect alumina and zirconia restorations because MDP monomer is directly linked to metal oxides and the phosphoric-acid groups of MDP can react with the oxide layer on the zirconia surface<sup>(4)</sup>.

Zirconia ceramics' clinical durability is influenced by numerous factors. These issues include the marginal fit and microleakage. The vertical marginal gap measures how far the restoration's margin is from the prepared finish line. Any marginal gap reveals the quality of the marginal adaptation and can affect how much leakage occurs, which may be one of the major reasons zirconia ceramic restorations fail.<sup>(5)</sup>

The null hypothesis of this study is that the treatment of fitting surface of zirconia with nanosilica may increase bond strength with resin cement and prepared crown. Also, it may improve internal adaptation but may decrease micro leakage.

## MATERIALS & METHODS

### A-Tooth selection

Twenty freshly extracted human maxillary first molar teeth were gathered from Minia University's Department of Oral and Maxillofacial Surgery. Carious lesions, coronal restorations, inadequate root formation, cracks, and perforations, or fractures were all exclusion criteria. Healthy, caries- and crack-free teeth with complete root development met the inclusion criteria. With a maximum dimension deviation of 10%, a digital calliper (Mitutoyo IP 65, Kawasaki, Japan) was utilised to

guarantee that the selected molars were all the same dimensions mesiodistal and buccolingual at the cemento-enamel junction. To remove any exterior plaque and depositions, all teeth were cleaned with an ultrasonic cleaner and preserved in saline (FIPCO, Egypt) at room temperature until usage.

### **B-preparation of epoxy mold**

The teeth were set in epoxy resin blocks as specimens using a custom-made cylinder mould with an inside diameter of 25 mm and a height of 10 mm. This was done to give the teeth support while the crowns were being prepared and tested. After filling the mould with the epoxy resin base and catalyst (CMB International, Giza, Egypt), teeth were inserted up to 2mm below the CEJ to allow for appropriate visibility of the crown margin during construction and final testing.

### **C-Tooth preparation for crowns:**

On greater control over the reduction amount, silicon index was manufactured on each tooth from hard duplication silicon material (Elite Double 22 Fast, Zhermack-Germany) before reduction.

Tooth preparation of a full-coverage crown was made with 1mm heavy chamfer finish line positioned 1mm above cemento enamel junction using Using a high-speed handpiece (NSK, Japan) paired with a surveyor (arm dental laboratory parallelometer A3005, Dentalfarm Srl, Italy) and water-cooling system, a full-coverage crown's tooth preparation was performed with a 1mm heavy chamfer finish line positioned 1mm above the cemento enamel junction. This standardised tooth preparation ensured a consistent degree of taper. Axial wall reduction (1–1.5mm) was accomplished using a taper stone with a round end (#845 Brassler, USA) and convergence of  $6 \pm 2$  degrees for each wall. Using a wheel diamond bur (Komet, USA), the occlusal surface was prepared while maintaining a vertical height of  $5 \pm 0.25$  mm.

### **D-Zirconia crowns fabrications by CAD/CAM**

A tiny layer of optical reflection spray (Occlutec Spray, Renfert Dental Corp., USA) was applied to each prepared tooth before optical impressions were taken using an extra oral scanner (Ceramill map400, Ammn Girrbach, Austria). Exocad design software was used to determine the tooth number, restoration type, and material for each zirconia crown. Certain characteristics, such as the margin definition, cement gap thickness (50 $\mu$ m) with 1 mm from the margin, and restoration thickness, were determined. The planned crown was delivered to the milling machine (Ceramill motion 2, Ammn Girrbach, Austria) after the tool path was calculated.

The set screw was used to fix the Ceramill white LT zirconia (Ammn Girrbach, Austria) in the spindle of the milling chamber of the milling machine (Ceramill motion 2, Ammn Girrbach, Austria). It took 25 minutes to mill. Crowns are white at this stage, indicating a partly crystalline condition.

Using a zirconia sintering furnace (Ceramill Therm 2, Amann Girbach AG) for 8 hours, all crowns were sintered. The temperature grew gradually until it reached 1450 degrees at a rate of 12 C/min, kept that temperature for an hour, and then slowly dropped five degrees per minute until it totally cooled. After sintering, all zirconia crowns were cleaned, and each one was checked to make sure it was properly set on the corresponding tooth.

### **Silica coating preparation:**

Controlled multiple-step hydrolysis of tetraethyl orthosilicate (TEOS; 98%, Sigma-Aldrich, USA) and cetyltrimethylammonium bromide (100g) (ACROS ORGANIS, Belgium). CTAB was produced in water medium with NH<sub>4</sub>OH (28–30%, Sigma-Aldrich, USA) as a catalyst to create a nonstructured silica-based sol concentration of 40%. In a dispersion medium (deionized water), one drop of produced silica sol was diffused<sup>(6)</sup>.

### **Nanosilica Coating of zirconia crowns.**

Three stages were taken to coat the sol-gel <sup>(6)</sup>:

1. Using a disposable brush, Silica Sol coatings were applied to the zirconia crowns' fitting surfaces (Meta Biomed, Chungcheongbuk, Korea).
2. To complete the sol to gel conversion, the coated crowns were stored in an incubator (POL-EKO-APARATURA, Poland) with 60% relative humidity for 24 hours.
3. After incubation for 30 min, they were heated in muffle furnace (Vulcan 3-550, USA) to 700C at rate of 10 C/min, and then cooled to room temperature.

### **Cementation Procedure:**

#### ***1- Surface Treatment of teeth:***

A 15-second application of etching gel (UNI-Etch, Meta Biomed) was followed by a 15-second wash drying period. Then, using a disposable brush (Meta Biomed, Chungcheongbuk, Korea), a coating of light-cured adhesive universal bonding agent (G-premio bond, GC, Tokyo, Japan) was applied. The light curing process took 20 seconds.

#### ***2- Surface Treatment of nanosilica coated crowns for cementation***

Using a disposable brush (Meta Biomed, Chungcheongbuk, Korea), a silane coupling agent (Bisco, USA) was applied to the fitting surface of crowns and allowed to dry for 30 seconds.

#### ***3- Cementation procedures of crowns***

TheraCem self-adhesive resin cement syringe (Bisco, USA) was used to cement all crowns in accordance with the manufacturer's instructions as follows: After cleaning, rinsing under running water, and drying, the teeth interface. After attaching the TheraCem auto mix tip, the cement was extruded into the fitting surface of the crown, immediately seated into the appropriate tooth, and maintained in

place using a specifically developed loading device. To avoid the crowns from rebounding during cementation, a constant weight of 5 kg parallel to the long axis of crown for 6 minutes. 2 seconds were used to light-cure the cement. With a sharp hand scaler, the excess cement was then scraped off the margins (Carl Martin, Pakistan). The cement was then given four minutes to set after each surface and margin light had cured for 20 seconds.

### **Aging of samples:**

In order to simulate a six-month clinical service period, samples of groups(thermocycling -nano silica with thermocycling) were aged through thermocycling (SD Mechatronik thermocycler, Germany) for 5000 cycles in tap water between 5°C and 55°C with a 30 second dwell time at each temperature and a 20 second transfer time.

### **- Microleakage test procedure:**

Nail polish varnish were covered the sample apical to the margin of the restoration by 1 mm to prevent any dye penetration except at the tooth/crown interface.

In order to test for microleakage, crowns were submerged in a 0.5% methylene blue solution for 24 hours. The samples were properly washed in water for five minutes after dye exposure. For holding purposes during sectioning, each sample was fixed in a PVC (polyvinyl chloride) tube with an internal diameter of 25 mm that had previously been filled with transparent acrylic resin (acrostone, Egypt). They were cut into bucco-lingual sections with an Isomet (4000 saw, Buehler, USA) using an 8-inch diamond disc with a 0.6-inch diameter. Dye penetration depth was measured using a light stereomicroscope (Nikon MA100 Japan) at 50× magnification and image analysis using computer software (Omnimet, Buchler, USA).

Statistical software (SPSS 19.0; SPSS Inc., Chicago IL) for Windows version 21 and Microsoft programmes were used to analyse the data on an

IBM compatible computer. For statistical analysis, one-way ANOVA and the Tukey test were employed. Means, standard deviations, medians, and ranges were measures used to represent quantitative data. If the P value was less than 0.05, it was deemed significant.

## RESULTS

Table (1) and figure (1) displays the means and standard deviations for the four study groups (control, thermocycling, nanosilica, and nanosilica-thermocycling). This study displayed that the nanosilica group (569.95 156.93  $\mu\text{m}$ ) and the nanosilica with thermocycling group (680.16 208.14  $\mu\text{m}$ ) had the lowest microleakage findings. The group that used thermocycling (891.21 72.33  $\mu\text{m}$ ) had the highest microleakage values.

1. There was no significant difference between the control and thermocycling groups. The mean value for the control group was (772.02 116.55  $\mu\text{m}$ ), whereas the mean value for the thermocycling group was (891.21 72.33  $\mu\text{m}$ ).
2. There was no significant difference between the nano silica group and the nano silica with thermocycling group. The mean value for the nano silica group was (569.95 156.93  $\mu\text{m}$ ), and the mean value for the nano silica with thermocycling group was (680.16 208.14  $\mu\text{m}$ ).
3. There was no significant difference between the control and nano silica groups.
4. Thermocycling group and nanosilica with thermocycling group there is no significant difference.

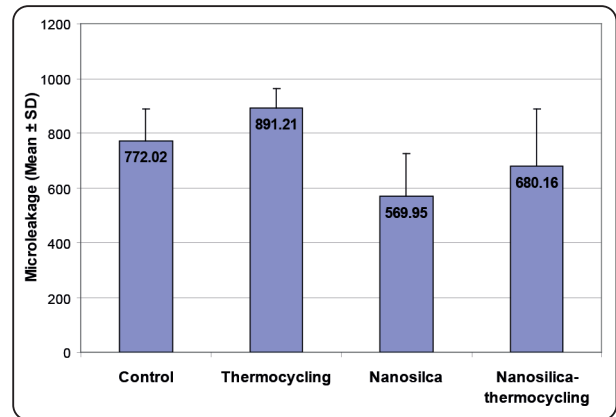


Fig. (1) Comparison between the four groups according to microleakage

## DISCUSSION

Dental ceramics based on zirconia offer greater mechanical qualities than other commercially available ceramic materials<sup>(7)</sup>. However, the bonding strength to resin cement has been observed to be weak due to the massive crystalline phase of zirconia-based ceramics, which makes traditional hydrofluoric acid etching treatment unfeasible. As a result, numerous researchers have researched different techniques of enhancing the adhesion between zirconia and resin cements in recent years<sup>(8)</sup>.

The most common fixed prosthodontic material, described by **Oilo et al., 2008 and Della Bona et al (2015)**<sup>(9)</sup> (1) owing to its higher strength, toughness, and superior long-term durability over other ceramics, was partly yttria-stabilized zirconia. <sup>(10)</sup>

Ceramill zi white LT zirconia (AMANNIR-RBACH AG, Austria) was chosen as one of the most widely utilised zirconia brands currently on the market.

TABLE (1) Comparison between the four studied groups according to Microleakage ( $\mu\text{m}$ )

Microleakage	Control	Thermocycling	Nanosilica	Nanosilica- thermocycling
Mean $\pm$ SD	772.02 $\pm$ 116.55	891.21 $\pm$ 72.33	569.95 $\pm$ 156.93	680.16 $\pm$ 208.14
Range	644.4-899.6	786.5-975.3	388.5-751.4	407.6-952.7

The degree of the marginal fit is crucial for the durability of the restoration and periodontal health since insufficient adaptation could lead to an increase in bacterial colonisation, secondary caries formation, and microleakage as a result of cement disintegration.<sup>(11)</sup> The gold standard for evaluating the efficacy of dental restorations is still clinical trials. However, the materials under examination can be outdated by the time valuable clinical data are available. Consequently, laboratory testing is still crucial for the assessment of dental materials.<sup>(12)</sup> The purpose of in vitro microleakage research is to evaluate the capacity of restorative materials to seal in vivo.<sup>(12)</sup>

Using a dye leakage model with methylene blue dye, which offers great precision because dye has no fillers, microleakage was examined in this work. Testing was done on the leakage at the die-cement interface since it has more biological significance.<sup>(13-15)</sup>

According to **Youngson, et al**<sup>(16)</sup> the greater potential of the pigment to enter dentin may be favourable since it may make it easier to distinguish between actual leakage and dentinal permeability. A significant complicating element in the measurement of penetration at the dentin/restoration contact is dye penetration into the dentinal tubules<sup>(17)</sup>. Testing for in-vitro microleakage that use dyes are thought to be stricter than those that use the oral cavity<sup>(18)</sup>. The following are some explanations given by **Pashley**<sup>(19)</sup> for the discrepancy between in-vitro and in-vivo leakage studies: 1) The dye diffuses more readily than bacteria and their byproducts; 2) The buildup of proteins and calcified debris may seal the marginal opening; 3) The positive pressure of the dentinal fluid in healthy teeth; and 4) The fibrinogen inside the sectioned tubules acts as a barrier to molecular penetration. Therefore, if a substance does well in the dye test, it will probably perform even better in a clinical setting.

Any in vitro study involving ceramic materials must include artificial ageing because it enables

the evaluation of restorations under realistically simulating clinical circumstances. Thermal ageing was recognised to impact restoration margin cement dissolving rates and marginal gap values.<sup>(20)</sup> All samples in this study underwent 5000 cycles of testing at 5° and 55°C water, which is equivalent to 6 months of clinical service.

Complex dense silica components can be produced using a sol-gel technique that produces high purity silica at a reasonably low cost without compromising quality. The production of the gel, drying, and densification are the three fundamental processes in the sol-gel process. In the current study, silicone alkoxide, such as TEOS, was combined with water to create pure silica. At room temperature, the alkoxide and water undergo a chemical reaction that creates a sol, which is a suspension of colloidal silica-based particles. The sol's viscosity is somewhat higher than that of water, making it easy to cover or shape into intricate patterns<sup>(6)</sup>.

Dip coating, meniscus coating, spin coating, and brush coating are some of the coating techniques used with nanostructured silica sol<sup>(21)</sup>. Brush coating was employed in our study.

The initial wet gel texture is strengthened in the second stage by an ageing and drying process. The dry gel can be heated further during the densification stage to increase the component's strength and thermal stability<sup>(6)</sup>. According to certain studies, the van der Waals force (electrostatic interaction) is what keeps silica coating on ceramics attached, and it needs to be appropriately heated to reach the desired strength<sup>(21, 22)</sup>. When the coating gel was heated in the air, the Si-OH groups reacted to create Si-O-Si (siloxane) linkages, which resulted in the creation of a silica network on the zirconia-fitting surface.

Meso-porous silica nanoparticles stand out among nanotechnological nanoparticles, according to **Carvalho G. et al. (2022)**<sup>(23)</sup> a system with significant biocompatibility, strong physical chemical stability, greater surface contact area, and

desirable and adjustable pore structure. Once they are developed and clearly defined, these pores can carry drugs and control their release. Surfactants are necessary to make this kind of nanoparticle, though, as they serve as pore templates. Cetyltrimethylammonium bromide (CTAB), a quaternary ammonium chemical that is frequently utilised as a surfactant in the creation of hollow mesoporous silica (HMSNs) and core-shell MSNs, is one of the most significant surfactants.

To help resin cement adhere to dental restorative materials, silane coupling agents like Bisco Silane are employed. They have two distinct functional groups: the -OR group and the >C=C vinyl double bond. The former can interact with the functional groups made up of >C=C bonds in resin cement. After hydrolysis, the latter transforms into -OH, which then chemically combines with the silica-coated zirconia surface<sup>(24)</sup>.

TheraCem self-adhesive resin cement (Bisco, USA) was used as the self-etch adhesive resin cement in this study. The reason for choosing it is that it contains MDP. Methacryloyloxyde Dihydrogen Phosphate monomers have been shown to be effective for sticking to non-silica-based polycrystalline zirconia materials in self-adhesive cements. Phosphate monomers are promising chemical agents for enhancing zirconia bonding, according to numerous research<sup>(25)</sup>. The self-etch method requires no additional etching or rinsing steps, which reduces the clinical application time and reduces the sensitivity of the method<sup>(26)</sup>.

In the present work, the impact of sol-gel ageing and silica coating on zirconia microleakage was assessed. The statistically significant difference in microleakage results between the groups was confirmed in the current investigation. The nanosilica group (569.95 156.93  $\mu\text{m}$ ) and the nano-silica with thermocycling group (680.16 208.14  $\mu\text{m}$ ) had the lowest microleakage findings, respectively. The thermocycling group showed the highest levels of microleakage among the groups (891.21 72.33 $\mu\text{m}$ ).

Clinically, microleakage happens when the adhesion process or adhesive contact becomes compromised, which may lead to additional adhesive failure. The findings of **Lung CYK et al (2013)**<sup>(3)</sup> regarding the impact of silica-coating by sol-gel method on resin-zirconia bonding can be used to explain this. They established that the zirconia's silica-coated layer and resin cement's interfacial failure caused the thermocycle group's even debonding. Therefore, it was evident that thermal fatigue and hydrolytic degradation during thermocycling had a negative impact on the bonding of resin to zirconia.

Microleakage before and after thermocycling was evaluated in the current investigation. It was observed that samples that had undergone thermocycling had more microleakage. The forces that arise during thermocycling may cause cracks to propagate at bonded surfaces, according to **kumar, et al (2018)**<sup>(13)</sup> and **Wahab, et al (2003)**<sup>(27)</sup>. The process of "percolation" is the inflow and outflow of oral fluids caused by changing gap dimensions once a gap has been created. Microleakage between the tooth and the restorative material may be seen when this gap develops after thermocycling. The largest clinically relevant stress will therefore occur from thermocycling the reconstructed tooth, which will also cause more microleakage.

According to **Rosentritt et al. (2004)**<sup>(28)</sup> resin cements and self-adhesive materials both exhibit strong marginal integrity and minimal microleakage, and the self-adhesive resin cements are potentially promising as an easily implementable resin cement substitute. On the other hand, glass-ionomer and resin-modified glass-ionomer cements should be taken into consideration as alternatives to resin cement, according to **Albert and El-Mowafy (2004)**<sup>(29)</sup> because of the distinction between the nature of dentinal adhesion, which is chemical (glass-ionomer) and micromechanical (resin). This can be explained by the cements' solubility, which is crucial to creating a better seal. Water-soluble cements, like glass-ionomer, degrade over time as a result

of thermocycling's harmful impacts. The insoluble resin cement, however, absorbs water, which might aid in reducing internal tensions brought on by polymerization shrinkage (**Eceyüksel, 2011**)<sup>(30)</sup>.

The results showed a strong positive correlation between internal fit and microleakage for all subgroups and in agreement with studies<sup>(31,32)</sup>.

According to **Sundar et al(2014)**<sup>(31)</sup> an increase in the marginal difference will proportionally result in more microleakage, which will raise the failure rate of a fixed dental prosthesis. **Hooshand et al(2011)**<sup>(33)</sup> observed no significant link between marginal fit and microleakage because of the wide range of leakage scores brought on by the use of many variables during testing and various laboratory methodologies.

## CONCLUSION

Within the limitation of this in vitro study, it is following conclusion can be drawn that:

1. Sol-gel processed nano-silica coating can decrease microleakage
2. Artificial aging has adverse effects on microleakage.

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