THE EFFECT OF TWO AGING METHODS ON THE FLEXURAL STRENGTH AND CRYSTAL STRUCTURE OF YTTRIA STABILISED ZIRCONIA POLYCRYSTALS (IN VITRO STUDY)

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ABSTRACT:

INTRODUCTION: Dental zirconia restorations present long-term clinical survival and stay in service within the oral environment for many years. However, low temperature degradation could affect their mechanical properties and survival.

Objectives: To investigate the effect of two aging methods on the flexural strength and crystal structure of yttrium-stabilized zirconia (Y-TZP).

MATERIALS AND METHODS: Thirty bar specimens were prepared from a Yttria stabilized zirconia polycrystals and were divided into 3 groups (control, aged for 720,000 mechanical loads of 50N and 3600 thermal cycles, aged for 1 hour using autoclave). The aging procedures represent 3 years of clinical use. The specimens were loaded until fracture and the crystalline phase polymorphs of the material (tetragonal, *t*, and monoclinic, *m*, zirconia) were investigated by x-ray diffraction (XRD). Further investigations were done using scanning electron microscope (SEM). Data was statistically analysed using ANOVA test.

RESULTS: Group B and C showed no statistical significance in their flexural strength with means of their break force (793.23±164.03) and (780.97 ± 257.25) respectively but statistically significant and higher than group A with mean (549.7 ± 54.14). The XRD showed nearly no change in the crystal structure between group A and B but an increase in the percent of monoclinic phase in group C. The SEM demonstrated a relatively homogenous size with particle size ranged between 400 to 570 µm for group A, while Group B and C showed an increase in particle size between 768 to 1150 µm respectively.

CONCLUSIONS: Both aging methods caused changes in the flexural strength and structure of the zirconia specimens with no significant difference between them.

KEYWORDS: Fixed prosthodontics, Zirconia, Aging, Low temperature degradation.

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INTRODUCTION:

Due to the patients need for esthetics and the presence of concerns about toxic and allergic reactions to certain alloys, patients and dentists have been looking for metal-free tooth-colored restorations. Therefore, the development of new high strength dental ceramics, which seem to be less brittle, limited in their tensile strength, and less subject to time dependent stress failure. These capabilities are highly attractive in prosthetic dentistry, where strength and esthetics are mandatory (1).

Many efforts were done to find all-ceramic materials with sufficient properties to enable their use for fixed partial dentures, which led to development of many new materials and processing techniques in the last decade (2).

Zirconia-based ceramic was introduced for dental use in 2002 and it became indicated for all-ceramic fixed partial dentures, and is gaining interest for other clinical applications and dental research (3).

The use of high-strength yttria-stabilized tetragonal zirconia polycrystals (Y-TZP) ceramics for fixed restorations has grown in recent years due to its high mechanical properties, and manufacturing convenience using CAD/CAM (computer aided designing/ computer aided manufacturing) procedures. Y-TZP has the most favorable properties with reasonable flexural strength, fracture resistance, and fracture toughness (4-6).

The main problem associated to the Y-TZP zirconia ceramics is their sensitivity to low temperature degradation (LTD) (7). Although this mechanism is very slow at oral temperatures other contributing factors such as constant humidity resulting from saliva, temperature, pH changes due to various drinks, repeated high occlusal loads due to mastication and autoclaving, can accelerate the aging process, and reduce the mechanical properties of the materials (8-9).

This study was an attempt to compare and evaluate the failure load and change in crystal structure of yttria stabilized zirconia polycrystals after aging with thermal cycling mechanical loading and in B class autoclave.

MATERIAL AND METHODS:

Master die preparation: A copper bar was fabricated with the dimensions of (20mm x 4mm x 2mm) then scanned after spraying it with anti glare-spray using 3D optical scanner.

Fabrication of test specimens: Milling of the zirconia specimens was done using Zirkonzahn m1 milling machine. A blank of partially sintered green state Prettau

anterior was clamped into milling chamber, using indentations at the blank margin.
Burs of 3 mm shank diameter, and different designs were used to mill the specimens. Three burs were used, 2L bur for rough milling, 1L bur for precise milling, 0.5 S for very precise milling.
Dry milling was done without water cooling. After specimens were milled, they were separated from the remaining of the blank using a bur mounted on straight handpiece, and smooth surfaces were attained.

The specimens were inserted into the sintering furnace, and the sintering cycle began by gradual rise in temperature, till reached 1500º C. At this temperature, the specimens spent holding time for two hours, followed by gradual decrease in temperature till room temperature.

Aging of the specimens: The specimens were divided into 3 groups where each group contained 10 zirconia bars and they were aged as follows:

Group A: 10 specimens as control group with no aging procedures.

Group B: 10 specimens were aged using thermal cycling mechanical loading for 720,000 mechanical loads of 50 N and 3600 thermal cycles corresponding to 3 years of clinical use (10). Thermocycling was done in a custom made thermocycling machine *. All specimens were cycled in hot and cold water baths between 5-55º C in 1 minute cycle for each bath, with a dwell time of 30 seconds.

Mechanical loading was performed in a custom made cyclic loading machine. Four metal spheres of 6 mm diameter were attached to the machine arm which was loaded by 5 Kg load. Four specimens were attached to metal rings that were attached by turn to the base of the machine. The 4 spheres were adjusted to be loaded at the centre of the bars. When the motor drive ran, the metal spheres moved up and down with a frequency of 2 HZ.

Group C: 10 specimens were aged for 1 hour under 2 bar pressure and 134°c inside an autoclave to resemble 3 years of clinical use (11).

Flexural strength test: The flexural strength test of the specimens of groups A, B, and C was measured by AGS-X 5 KN Shimadzu universal testing machine equipped with a 10-kN load cell and crosshead speed of 1 mm/min according to ISO 6872 until fracture. Mean values of flexural strength were calculated using the following equation:

$$\sigma = \frac{3PL}{2\,wb^2}$$

Figure 1: Mechanical loading machine.

Where P is the breaking load in Newtons; L is the test span (center-to-center distance between support rollers) in millimeters, w is the width of the specimen in millimeters; b is the thickness of the specimen in millimeters. Each specimen was attached with springs to the base of the machine and a knife edge indenter was attached by means of a stylus to the upper compartment of the machine.

Crosshead speed of 1mm/min was chosen. After the indenter was adjusted manually through a manual panel, stress and strain were adjusted to be zero newton, and zero mm respectively.

Afterwards, the operation was started, and the load was raised gradually, until sudden sharp decrease of the force, which was also accompanied by fracture of the specimens. The maximum load before the sharp decrease of force was recognized as the "flexural strength", and was determined for each specimen in newton.

Crystal structure: X-Ray 7000 Shimadzu X-ray diffractometer was used with a representative specimen of each group to investigate phase composition using X-ray diffraction (XRD).

The X-ray diffractometer operating with CuKα radiation (λ=0.154060nm) generated at 30kv and 30mA. Scans were performed at $2°min^{-1}$ for 2Θ values between 20° and 80° where Θ is the angle of reflection, which covers the positions of the highest peaks of tetragonal and monoclinic phases of zirconia.

The crystalline size (D) in nm was calculated from the reflection of tetragonal zirconia phase at (101) peak, and of monoclinic phase at (-111) peak using the Schererr relationship.

$$D = \frac{k\lambda}{\beta \cos \Theta}$$

Where k is the crystalline shape constant (0.9), λ is the radiation wavelength (A°), βis the line breadth (radians), and Θ is the Bragg angle.

The relative amount of monoclinic phase (X_m) was calculated by the method of Gravie and Nicholson, which is most commonly applied to determine the phase composition of zirconia powders and compacts.

$$X_m = [I_{m(-111)} + I_{m(111)}]/[I_{m(-111)} + I_{m(111)} + I_{t(101)}]$$

Where I_t and I_m represent the integrated intensity of the tetragonal (101) and monoclinic [(111),(-111)] peaks around 30°, 31° and 28° respectively.

Scanning electron microscope: JEOL JSM 636OLA Analytical Scanning Electron scanning electron microscope was used with a representative specimen of each group to be further analysed. Selected specimens were gold coated, and introduced to the vacuum chamber. Areas of interest were captured and recorded.

Statistical analysis: Data were fed to the computer and analyzed using IBM SPSS software package version 20.0**. (**Armonk, NY: IBM Corp**)** The Kolmogorov-Smirnov test was used to verify the normality of distribution Quantitative data were described using range (minimum and maximum), mean, standard deviation and median. Significance of the obtained results was judged at the 5% level. The used tests was F-test (ANOVA) for normally distributed quantitative variables, to compare between more than two groups, and Post Hoc test (LSD) for pairwise comparisons.

RESULTS:

Group B and C showed no statistical significance in their flexural strength with means of their break force (793.23±164.03) and (780.97 ± 257.25) respectively but statistically significant and higher than group A with mean (549.7 ± 54.14). The flexural strength test results were shown in table 1.

Table 1: Comparison between study groups according to force.

Force	Group I (n= 10)	Group II (n= 10)	Group III (n= 10)	F	p
Max					
Min. – Max.	461.56 – 629.84	669.06 – 1254.38	532.34 – 1284.84		
Mean ± SD.	556.64 ± 54.01	802.33 ± 166.11	799.92 ± 242.94	6.677*	0.004*
Median	570.08	760.70	738.98		
Sig. bet. Grps.	p_1= 0.004*,p_2= 0.004*,p_3= 0.975				
Break					
Min. – Max.	461.56 – 629.84	669.06 – 1241.56	489.22 – 1283.91		
Mean ± SD.	549.7 ± 54.14	793.23 ± 164.03	780.97 ± 257.25	5.881*	0.008*
Median	558.67	745.94	703.28		
Sig. bet. Grps.	p_1= 0.005*,p_2= 0.007*,p_3= 0.879				

The XRD showed no change in the crystal structure between group A and B but an increase in the percent of monoclinic phase in group C. The XRD of groups A, B and C are shown in figures 2, 3 and 4 respectively.

The SEM demonstrated a relatively homogenous size with particle size ranged between 400 to 570 µm for group A, while Group B and C showed an increase in particle size between 768 to 1150 µm respectively.

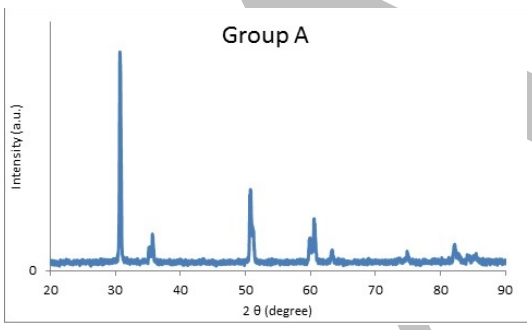

Figure 2: The result of XRD of group A.

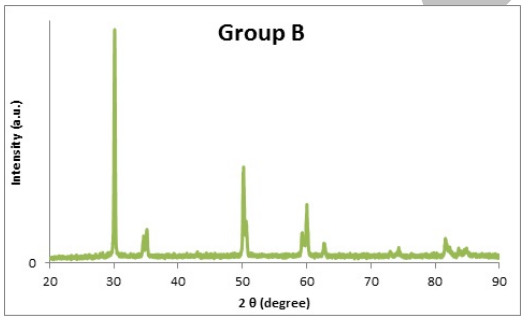

Figure 3: The result of XRD of group B.

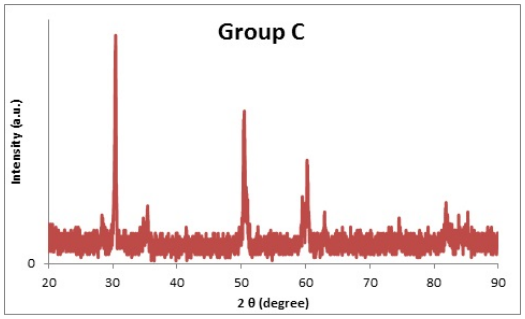

Figure 4: The result of XRD of group C.

DISCUSSION:

This in-vitro study was an attempt to evaluate the effect of two different aging methods on the flexural strength and crystal structure of yettria stabilized zirconia polycrystals.

Due to uncertainty of determining activation energy of t-m transformation (12) data from such tests can be extrapolated anywhere between 1 and 12 years. Chevalier et al (11) suggested that 1 hour in autoclave is equivalent to 3-4 years low temperature degradation at 37°c assuming comparable time-temperature equivalences less than 25% t-m transformation thus remaining below acceptable amount according to ISO standard 133356:2008. Rosentritt et al (10) suggested that 720,000 cyclic loads and 3600 thermal cycles using thermal cyclic mechanical loading corresponds to 3 years of clinical use.

Thermocycling was chosen because it simulates intraoral temperature changes and chewing forces. Aging was done to correspond 3 years of clinical use. During the cyclic loading , sphere indenters with a diameter of 6 mm were used as antagonists in this study according to previous investigations done by Zou et al (13), Buer et al (14) and Albrecht et al (15).

Flexural strength can be defined as the final force needed to cause fracture of a fragile material and is strongly affected by the defect and flaw present in the materials so it is a very important mechanical property to predict the performance of a material (16). Three point bending test is the standard method to be used to measure the flexural strength of all ceramic materials so it was used in this study.

The result of flexural strength of group B was significantly higher than the control group A this may be contributed to the increased percent of monoclinic phase which leads to volume expansion thus diminishing residual stresses and inhibiting crack growth thus increasing the flexural strength (17).

The result of this study was in agreement with kim et al (18).

The result of this study was in disagreement with Papanagiotou et al (19), Ardlin (3), Yilmaz et al (20), where there was no change in the flexural strength after aging and this may be contributed to the longer period used in this study

XRD was used to investigate the increase of monoclinic phase after aging since the difference between tetragonal and monoclinic phases is crystallographic. Major contributions by Garvie and Nicholson were made for the use of XRD to differentiate between tetragonal an monoclinic phases (21).

The results of XRD showed that nearly there was no significance of monoclinic phase this may be due to the change was not enough to appear on the XRD or maybe the investigated part of the specimen was not affected by low temperature degradation yet.

The result of this study was in disagreement with Ardlin (3) and Kim et al (18).

The SEM results of this study showed an increase in particle size of the aged specimens which indicates the transformation of crystals polymorphs from tetragonal to monoclinic which is larger in size.

Autoclave was chosen because low temperature aging of zirconia is usually done in autoclave where temperature, pressure of water vapor and elapsed time are controlled experimental variables (22). Aging was done to correspond 3 years of clinical use.

After aging procedure flexural strength increased than the control group. These results are in agreement with Garvie and Nicholson (21), Kobayashi et al (23), Chevalier et al (10) Amaral et al (24) and Pereira et al (25).

On the contrary from group B there was an increase in the percent of monoclinic phase in the specimen that was aged using autoclave and this may be contributed to the high activation energy of the autoclave. The results are in agreement with kawai et al (26) Tholey et al (27) Kosmac et al (28), li et al (29), Yasmine et al (30) and Tutku et al (31).

The SEM results of this study showed an increase in particle size of the aged specimens which indicates the transformation of crystals polymorphs from tetragonal to monoclinic which is larger in size.

Since low temperature degradation proceeds at a slow pace, to our knowledge no data is currently available at the body temperature. Life time predictions are based on accelerated aging tests.

CONCLUSIONS:

It was concluded from this study that:

1) Both methods of aging increased the flexural strength.
2) Both methods caused t-m transformation.
3) Autoclave can be used in aging zirconia specimens.

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CONFLICT OF INTEREST:

The authors declare that they have no conflicts of interest.

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