Effect of Autoclaving on some mechanical properties of dental machined zirconia for crown restorations

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Abstract : Yttrium-stabilized tetragonal zirconia polycrystal (Y-TZP) has been introduced into the field of dentistry as dental crowns and implant prosthesis due to superior strength and toughness compared to other dental ceramic systems, by using computer programs for design and manufacture. Before implantation, the use of autoclave is mandatory for sterilization and so is the study of its effect on the mechanical properties. **PURPOSE:** The objective was to study the effect of autoclaving on the mechanical properties of dental monolithic zirconia. **MATERIALS AND METHODS:** Y-TZP blocks of Metoxit Z-CAD® HTL (Meoxit AG, Thayngen, Switzerland) were used in this study. The partially sintered blocks were cut into discs with the size of 10 mm (diameter) × 2 mm (thickness), then fully sintered at 1550℃ according to a cycle recommended by the manufacturer in a sintering furnace (Luoyang, China), for 5 hours. Specimens were sterilized in an autoclave at a temperature of 134 ^{o}C for 10 hours. The specimens were divided into two groups; sintered as the control group and autoclaved. The mechanical properties of both groups were measured; hardness by using Vickers hardness tester (MVK-H2, China), Surface roughness measurement by using surface profilometer (Surfcom 130A, Tokyo Seimitsu, Tokyo, Japan), Surface topography analysis by using scanning electron microscope and surface chemistry analysis by using Energy dispersive spectroscope (JSM-6360, JEOL, Tokyo, Japan), X-ray diffraction analysis by using diffractometer (DMAX 2500; Rigaku, Tokyo, Japan), Fracture resistance test by manufacturing crowns and using Instron(Nikon, Tokyo, Japan), The statistical analysis was done using(IBM SPSS Statistics 26). **RESULTS:** The control group hardness is higher than the autoclave group, XRD test shows that tetragonal phase is stable. Crown Fracture testing showed that the fracture resistance of the monolithic zirconia crowns increased (mean rank=24.00) when subjected to autoclaving treatment of 134 ^{o} C for 10h,than that of the control group (mean rank=8.5)

Key words Zirconia , low temperture degdration, fracture resistance.

INTRODUCTION

The increased popularity of all-ceramic materials as an alternative to metal ceramic restorations, is attributed to their excellent aesthetics, chemical stability, and biocompatibility. The ceramic zirconia has been used as a biomaterial for implants and dental crowns because it has good mechanical properties; very similar to those of metals and its color is similar to tooth color, good wear resistance and friction, superior toughness, fatigue resistance and excellent biocompatibility [1,2].

Zirconia has a polymorph form because has three crystal structure monoclinic, tetragonal and cubic phases, at room temperature zirconia naturally is in its monoclinic phase. The phase material is changes to tetragonal phase when temperature is 1170° C, whenever increase of temperature to 2370° C obtain the cubic phase.

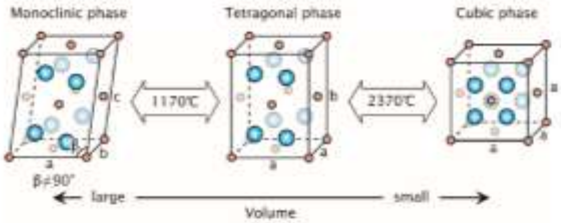

Figure 1. Schematic of temperature-dependent crystalline structure of zirconia. Red spheres = Zr, Blue spheres = O. The figure is modified from (Hannink et al., 2000; Amarante, 2018b).

The metastable tetragonal phase in zirconia is useful in application of the material and resistance against crack propagation. When zirconia is exposed to stress, the metastable tetragonal phase transforms to monoclinic phase [3]. Stress induced phase transformation occurs, increasing its crack propagation resistance. This process involves compressive stress generation around micro-cracks [4], and results in arrested crack propagation. Zirconia (zirconium dioxide, ZrO2) with higher strength (900-1200 MPa) [5,6,7] has been applied frequently.

The development of computer aided design/computer aided manufacturing (CAD/CAM) technology [8;9] has made zirconia popular in dentistry. Furthermore, the translucent tooth-colored zirconia ceramics, monolithic zirconia restorations without veneering material, also called full-contour zirconia has been also developed [10].

The metastable tetragonal phase spontaneously transforms into the monoclinic phase in a humid atmosphere even without mechanical stress, which begins at the surface and enters the bulk of the material. This process is often referred to as low-temperature degradation (LTD) or aging [11]. The objective of this study was to find the effect of autoclaving on the mechanical properties of dental monolithic zirconia implant and crown restorations.

MATERIAL AND METHODS:

Preparation of specimens

Y-TZP blocks of (Metox, Switzerland) are partially sintered. The blocks were cut into discs with the size of 10 (diameter) × 2mm (thickness) approximately. Then the specimens were fully sintered at 1550℃ according to a cycle recommended by the manufacturer in a sintering furnace (Luoyang, China), for 5 hours. The specimens were divided into 2 groups, 10 specimens for each. All specimens were manually polished with a rubber polishing cup, for 5 minutes. The purposes of the polishing were to remove all surface residual stresses, to round chipped corners of the specimens, and to eliminate any cracks from inherent material defects.

Low temperature aging

Specimens were artificially aged in distilled water by heat-treatment at a temperature of 134^{o} C for 10 hours, the Autoclave reactor (ECO solution, Seoul, Korea) was used in order to induce the phase transformation at the surface.

Surface roughness measurement

Surface roughness values were measured by a profilometer (SURFTEST SJ-201, Mitutoyo Corp., Kawasaki, Japan). Ten specimens per group were measured for surface roughness.

The stylus moved back and forth across each specimen, five readings were recorded, and the mean roughness value (Ra) of the specimen was calculated. The cut-off length was 0.8 mm, at 0.5 mm/s scanning speed, the stylus with a tip diameter of 2.5 μm. The resolution of the recorded data was 0.01 μm.

Surface hardness measurement

Surface hardness were measured for ten specimens from each group by the micro-Vickers hardness test. Indentation was produced on the polished surface under a load of 9.8 N for 15 s using a Micro hardness tester Model HV-1000 (JINAN PRCISION TESTING EQUIPMENT CO., LTD, China).

Surface analysis (Topography and elemental composition analysis)

The surface morphology of each treated group was assessed using a scanning electron microscope (SEM, JSM-6510LV, JEOL, Tokyo, Japan). For this purpose, two specimens from each group were produced as previously mentioned, cleaned in an ultrasonic bath with 96% ethanol for 2 min and then air dried. Afterward, specimens were affixed on metallic stubs, gold sputter-coated (SPI-MODULETM, SPI Supplies, USA), and examined under a SEM to detect topography of the treated surfaces (X 5000 and X 20,000). Energy Dispersive Spectroscope (EDS) was employed to investigate the elemental composition of the Zirconia specimens. The working distance and voltage used was 15 mm and 20 V respectively.

X-ray diffraction analysis

To determine the crystalline phase fraction, X-ray diffraction analysis was carried out using XRD device (Siemens (Germany) D5000 powder diffractometer). The specimens were placed in the holder of a diffractometer and Cu K_α radiation wavelength = 0.15406 nm, with a nickel filter at 40 kV and 30 mA was applied. The diffractometer was operated within the range of $10^o < 2^o < 100^o$ using step size of 0.05 deg./sec.

Fracture resistance test:

In this laboratory test, a standard die was prepared to achieve an occluso-gingival length of 7.5mm, a diameter of 7mm, a finish line of 1.5mm and a taper of 6° at each side. The finish line design was the radial shoulder. The die impression was made using silicone impression material (Speedex, Coltene/Whaledent, Switzerland) and was poured with epoxy resin (GC Pattern Resin, Tokyo, Japan). The main die was scanned with the Cercon Eye® scanner (Cercon, DeguDent, Hanau, Germany), and a three-dimensional model of the die was fabricated; the thickness of the cement space was considered to be 30μm. Twenty- zirconia crowns (Metoxit, Switzerland) were made of pre-sintered zirconia blocks using the data obtained by scanning the die and produced using CAD/CAM technology (Arum x5 400, Doowon Co., Ltd. Daejeon, South Korea). The final sintering of the zirconia crowns was carried out in the Cercon sintering machine (Luoyang, China) at 1550°C for 5 hours.

Zinc phosphate cement was used to cement the crowns to the dies. The crowns were then seated on the corresponding dies with a mild finger pressure, and a constant pressure of 15N was applied on the specimens using a 0.5kg weight via an acrylic connector fabricated equal to the size of the crowns. The excess cement was removed by a hand instrument after setting. The zirconia copings were divided into two groups: Control (n=10), and Autoclave treated (n=10). The point of load application, at the center of the occlusal surface, was marked on the specimens which were placed in the universal testing machine (Instron Universal testing machine, 3345, USA) at a cross head speed 5mm/min, with a round tip of 4 mm diameter.The fracture load was recorded when the catastrophic fracture occurred, and the Weibull plot curves and distribution analysis were done.

RESULTS:

Hardness

The statistical analysis of the data was carried out using IBM SPSS Statistics 26. The independent variable is the autoclave treatment method applied to specimens. The independent variable includes two groups (two types of treatment): ‘Before Autoclaving’ and ‘After Autoclaving’.
The dependent variable is the hardness value measured.

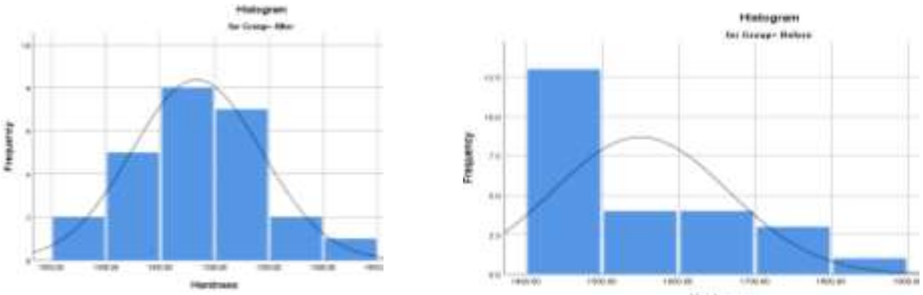

Figure (1) Histogram of the hardness values for the two groups.

Mann-Whitney Test:

Test Statistics

	Hardness
Mann-Whitney U	199.500
Wilcoxon W	524.500
Z	-2.198
Asymp. Sig. (2-tailed)	.028
Exact Sig. (2-tailed)	.027
Exact Sig. (1-tailed)	.014
Point Probability	.000

The results show that there is a significance difference ($p = 0.027$) between the hardness values measured of the specimens in the control group and those in the treatment group. The hardness values of the control group (mean rank = 30.02) is higher than the Autoclave treated group (mean rank = 20.98); see Figure 2.

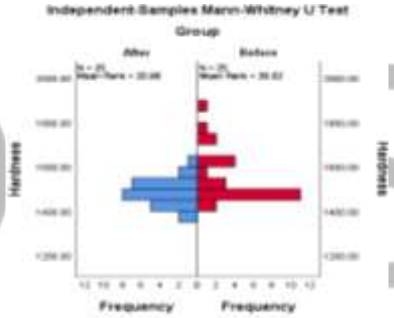

Figure (2) Comparing the ranks of the two groups

Roughness

The statistical analysis of the date was carried out using IBM SPSS Statistics 26. The independent variable is the treatment method applied to the tooth crown. The independent variable includes two groups (two types of treatment): 'Before Autoclaving' and 'After Autoclaving'. The dependent variable is the roughness value measured.

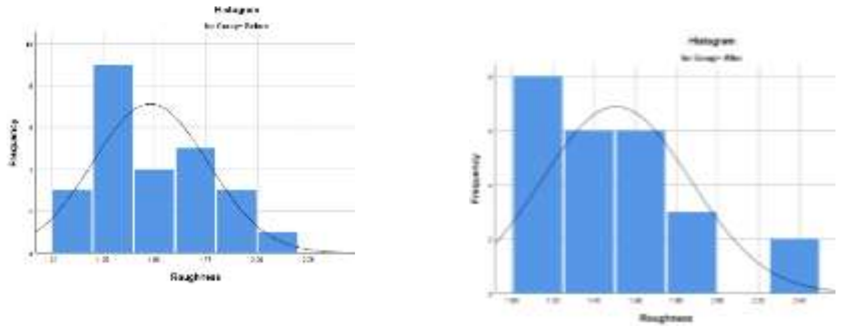

Figure (3) Histogram of the roughness values for the two group

Mann-Whitney Test:

Test Statistics

	Roughness
Mann-Whitney U	308.500
Wilcoxon W	633.500
Z	-.078
Asymp. Sig. (2-tailed)	.938
Exact Sig. (2-tailed)	.943
Exact Sig. (1-tailed)	.471
Point Probability	.004

The results show that there is no significance difference ($p = 0.943$) between the roughness values measured of the specimens in the control group and those in the autoclave treated group. The two groups have near equal mean rank (25.66 and 25.34); see Figure 4.

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Figure (4) Comparing the ranks of the two groups Fracture resistance:

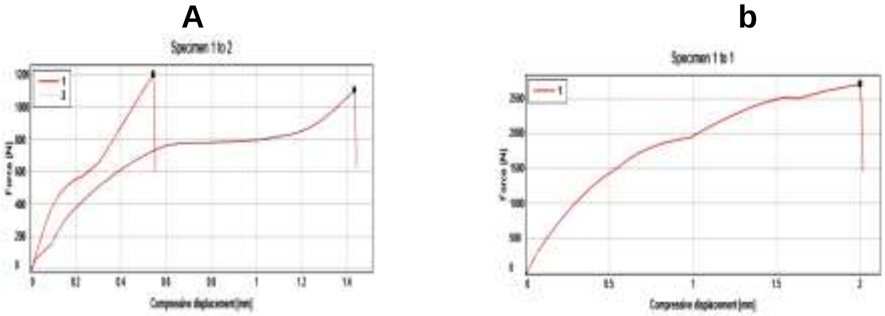


Figure (5) is representative (a)compressive stress for control group, (b) compressive stress for autoclave group

Statistical Analysis (Compression Test):

Goodness-of-Fit Test for the Weibull Distribution:

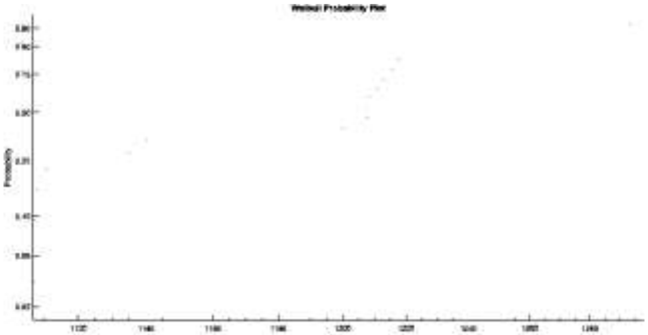


Figure (6) Weibull probability plot for the Control group

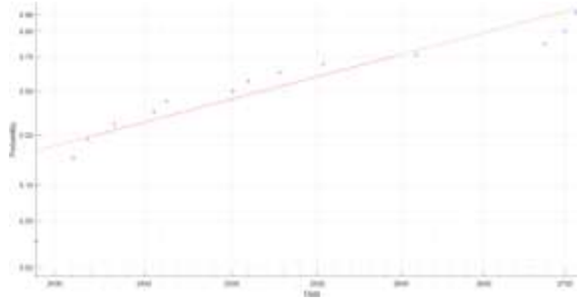


Figure (7) Weibull probability plot for the Autoclave group

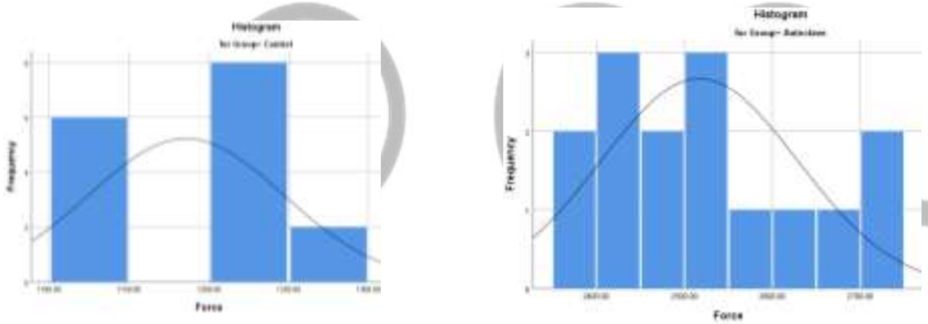


Figure (8) shows the histogram of the fracture resistance test values measured for the two groups (Control and Autoclave). It is apparent that the data from the two groups heavily skewed.

The Shapiro-Wilk test was used for testing the normality of the data. For the Control group yielded a statistical significance of $p = 0.30$, and for the Autoclave group of $p = 0.064$. The null hypothesis is rejected in both cases and the data is assumed to be non-normal.

The values of the Autoclave group (mean rank = 24.00) is higher than the Control group (mean rank = 8.5); see Figure (9).

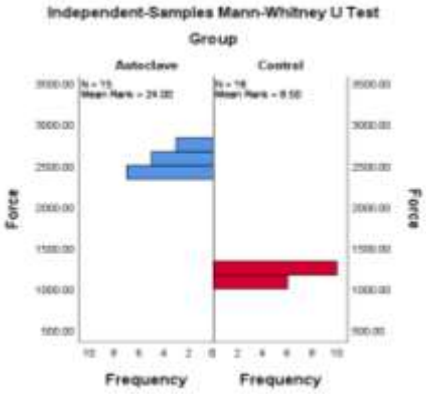
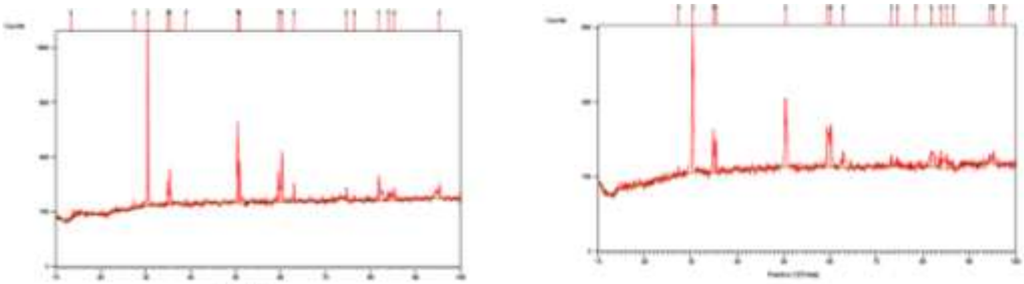


Figure (9) Comparing the ranks of the two groups

X-ray diffraction analysis:

XRD analysis of sintered Y-TZP ceramics indicate that the control group consisted of tetragonal zirconia peaks. After autoclave treatments, detectable monoclinic peaks appeared in the XRD pattern.

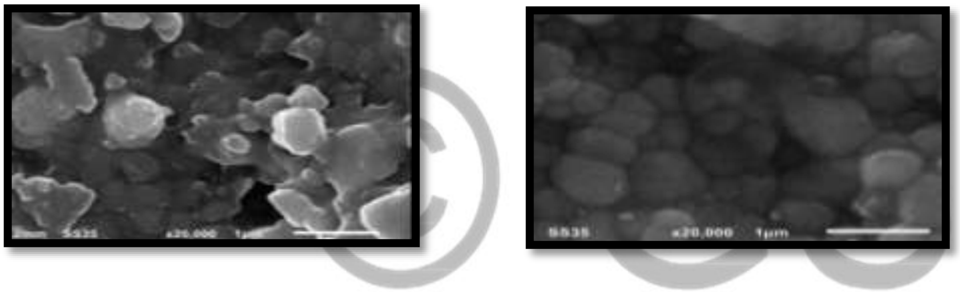
<u>Additional pattern</u>:



A. B.

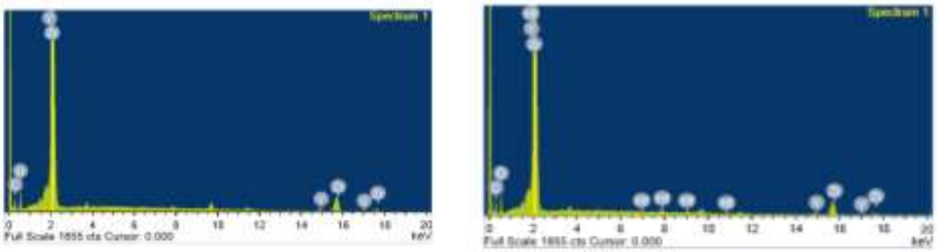
Figure (10)reprehensive (A)control group (B)autoclave group Surface analysis (Topography analysis):

It is clear that there are differences between the two groups in terms of porosity in the control group(a) we observed that the granules are more spaced, which indicates that porosity is high. While in the group autoclave(b) note that the stacking between the granules are more than in the other group, as it is more coherent with less interstitial spaces.



A B

Figure(11)The images taken by scanning electron microscope, for control group(a)&group autoclave(b)with 134 ^{o}C for 10h.

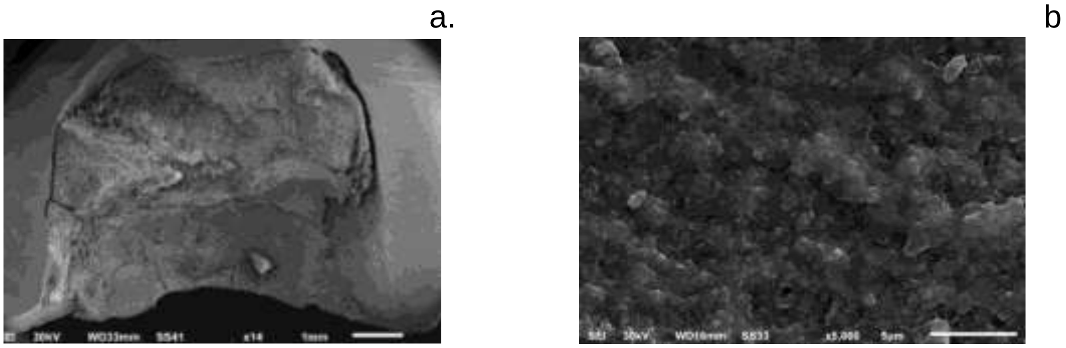


a b

Figure(12) is represented Elemental analysis in Weight% Atomic% for (a)control group,(b)autoclave group

It could be seen from the EDX that both control and autoclave groups mainly consists of Zirconia, Yttrian, Hafnia, oxygen and carbon.

SEM analysis of fractured monolithic zirconia crown:



Figures (13)are representative micrograph and SEM image of fractured monolithic zirconia crown for control group without autoclaving treatment.

Figures (14) are representative micrograph and SEM image of fractured monolithic zirconia crown for autoclave treatment group at 134 c for 10 h.

Discussion:

Due to the normal color of Y-TZP (*i.e.* bright white), its application in prosthetic dentistry was limited to implant abutments or frameworks of prostheses until recently, the development of translucent tooth-colored zirconia, enables the fabrication of monolithic zirconia restorations without veneering material, also referred to as full-contour zirconia restorations [12]. Thus, the demand for tooth-colored zirconia ceramics is increasing. Tooth-like color can be given to zirconia by adding coloring pigments, such as metal oxides [13,14].

As a result of utilizing the zirconia ceramics for the fabrication of tooth-supported restorations, this encouraged the clinicians to extend its application for implant-supported restorations. Utilizing zirconia as implant-supported restorations is due to the higher toughness and the lower modulus of elasticity of zirconia. In stabilized and transformation toughened forms, zirconia provides some advantages over alumina in order to solve the problem of alumina brittleness and the consequent potential failure of implants [15].

The current study found that Low–temperature aging in autoclaving did not prompt a negative effect on the mechanical properties and structural reliability (Weibull modulus) of material, the autoclave treatment did not affect the surface roughness, and this is similar to a study by Nakamura, et al, 2015 [16], who suggested that the surface roughness was not affect by LTD, because the tip diameter of the stylus (2.5 μm) that was used is much larger than the zirconia grains.

The hardness results value showed that the control group is higher than autoclave group, that is similar with other previous study that assumed that the Vickers Hardness of Y-TZP decreased with autoclaving time. This could be attributed to the induced LTD, as it generates micro-cracks on the material surface which decreases surface hardness. [16,17]

The SEM results of the autoclave group shows homogeneous grainy structure, on the other hand, the control group shows many grooves and voids. This may be due to the effect of Autoclave treatment that make rapid modifications in grain size, this Means that each grain was pushed out of the surface and roughness of surface increased due to low Temperature aging.

XRD test showed that the Autoclaving led to some crystallographic transformation to monoclinic, when the specimen was subjected to autoclaving for 10 hours. The monoclinic phase on the surface was detected by XRD, but in some studies the monoclinic phase was observed in autoclave after somehow longer duration of time (50h,100h) in Nakmura, et al, 2015 [16]. This crystallographic transformation might be initiated when a nucleus is first formed at a specific grain that is more susceptible to the phase transformation because of a disequilibrium state, such as large grain size, lower content of stabilizer and the presence of residual stresses [18]. Although the mechanism of the phase transformation caused by water molecules has not been fully elucidated [19], following steps are proposed [20,21,22]:

1) Chemical adsorption of H_2O on ZrO_2 surfaces
2) Formation of Zr-OH bond disrupting Zr-O-Zr bond
3) Penetration of OH^- and/or O^{2-} into the inner part by grain boundary diffusion
4) Filling of oxygen vacancies by OH^- and/or O^{2-}
5) Reduction of the oxygen vacancies destabilizing tetragonal phase

It has been established that the stability of tetragonal phase, and in turn susceptibility to LTD, depends on several material properties, such as density, purity, grain size, and type and content of stabilizer [23,11,19].

CONCLUTION:

Based on the results of this study, and despite some limitations, we can conclude that; autoclaving treatment did affect the mechanical properties of Y-TZP ceramics. When applying aging temperature at 134^{o}C for 10h, surface hardness decreased. Autoclaving treatment affected microstructure of the material, as the grains became more homogeneous. Autoclaving treatment proved progress in the crowns fractural; resistance as the monolithic zirconia crowns seem to have sufficient strength even when assuming LTD occurs.

REFERENCES:

1-Craig RG, Powers JM (2002). 'Restorative Dental Materials', Mosby Inc, St. Louis.

2-van Noort R (2002). 'Introduction to dental materials.', Mosby, London

3- Hannink, R.H.J., Kelly, P.M., Muddle, B.C. (2000) Transformation toughening in zirconia-containing ceramics. *J Am Ceram Soc* 83*:*461-487

4- Kelly, P.M., Ball, C.J. (1986) Crystallography of stress-induced martensitic transformations in partially-stabilized zirconia. *J Am Ceram Soc* 69*:*259 264.

5- Christel, P., Meunier, A., Heller, M., Torre, J.P., Peille, C.N. (1989)Mechanical-properties and short-term invivo evaluation of yttrium-oxidepartially- stabilized zirconia. *J Biomed Mater Res* 23*:*45-61.

6- Kosmac, T., Oblak, C., Jevnikar, P., Funduk, N., Marion, L. (1999) The effect of surface grinding and sandblasting on flexural strength and reliability of Y-TZP zirconia ceramic. *Dent Mater* 15*:*426-433.

7- Guazzato, M., Quach, L., Albakry, M., Swain, M.V. (2005) Influence of surface and heat treatments on the flexural strength of Y-TZP dental ceramic. *J Dent* 33*:*9-18.

8- Manicone, P.F., Rossi Iommetti, P., Raffaelli, L. (2007) An overview of zirconia ceramics: Basic properties and clinical applications. *J Dent* 35*:*819-826. Denry, I., Kelly, J.R. (2008) State of the art of zirconia for dental applications. *Dent Mater* 24*:*299-307.

9- Denry IL, Kelly JR (2008) State of the art of zirconia for dental applications.
Dental Materials 24, 299-307

10- Christensen, G.J. (2011) The all-ceramic restoration dilemma: Where are we? *J Am Dent Assoc* 142*:*668-671

11-Chevalier, J., Gremillard, L., Deville, S. (2007) Low-temperature degradation of zirconia and implications for biomedical implants. AnnRev Mater Res 37:1-32.

12-Cales, B. (1998) Colored zirconia ceramics for dental applications. Bioceramics 11:591-594.

13- Shah, K., Holloway, J.A., Denry, I.L. (2008) Effect of coloring with various metal oxides on the microstructure, color, and flexural strength of 3Y-TZP *J Biomed Mater Res B Appl Biomater* 87*:*329-337

14- Hjerppe, J., Narhi, T., Froberg, K., Vallittu, P.K., Lassila, L.V. (2008) Effect of shading the zirconia framework on biaxial strength and surface microhardness. Acta Odontol Scand 66:262-267.

15 - Yildirim M, Fischer H, Marx R, et al. In vivo fracture resistance of implant supported all-ceramic restorations. J Prosthet Dent 2003; 90: 325-31.

16- Keisuke Nakamura;(2015)..mechanical and microstructural properties of monolithic zirconia crown frature resistance and impact of low-temperture degradation.

17-Elshazly, E.S., El-Hout, S.M., Ali, M.E.S. (2011) Yttria tetragonal
zirconia biomaterials: Kinetic investigation. *J Mater Sci Technol* 27*:*332- 337

18- Chevalier, J. (2006) What future for zirconia as a biomaterial? Biomaterials 27:535-543.

19- Lughi, V., Sergo, V. (2010) Low temperature degradation -aging- of zirconia: A critical review of the relevant aspects in dentistry. *Dent Mater* 26*:*807-820

20- Yoshimura, M., Noma, T., Kawabata, K., Somiya, S. (1987) Role of H2O
on the degradation process of Y-TZP. J Mater Sci Lett 6:465-467.

21 - Lawson, S. (1995) Environmental degradation of zirconia ceramics. J Eur Ceram Soc 15:485-502.

22-Chevalier, J., Gremillard, L., Virkar, A.V., Clarke, D.R. (2009) The tetragonal-monoclinic transformation in zirconia: Lessons learned and future trends. *J Am Ceram Soc* 92*:*1901-1920

23- Clarke, I.C., Manaka, M., Green, D.D., Williams, P., Pezzotti, G., Kim, Y.H. et al. (2003) Current status of zirconia used in total hip implants. J Bone Joint Surg Am 85-A Suppl 4:73-84.