Original Article / Özgün Araştırma

Effect of Low-thermal Degradation on the Flexural Strength of Y-TZP Ceramics

Düşük Isı Bozunmasının Y-TZP Seramiklerinin Eğilme Dayanımına Etkisi

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Abstract

Objective: This research aimed to look into the aging characteristics of three distinct yttria-stabilized tetragonal (t) zirconia polycrystalline, as well as the impact of low-thermal degradation on flexural strength.

Materials and Methods: One hundred twenty disc-shaped specimens were obtained from three different brands (Supra Zr, Zirkonzahn Prettau and CopraPretty). For each brand, specimens in the control groups were left in distilled water at 37 °C for 5 h whereas those in experimental groups were artificially aged under three different conditions. In the thermal cycle group, specimens were fatigued for 10,000 between baths held at 5 °C and 55 °C; left in 4% acetic acid at 80 °C for 168 h in acetic acid groups, and aged at 134 °C, 0.2 MPa for 5 h in autoclave aging groups. T to monoclinic phase transformation (Xm) quantified using X-ray diffraction for all groups. One randomly selected specimen surface was analyzed by scanning electron microscopy from each subgroup. The biaxial flexural strength test was used to assess the flexural strength of the specimens. The data were statistically analyzed.

Results: The highest relative amount of the Xm content was obtained in the autoclave aging groups (Supra Zr 0.36±0.01, Zirkonzahn Prettau 0.41±0.02, CopraPretty 0.58±0.04). There was no significant difference between Zirconzahn Prettau and Supra Zr materials ($p>0.05$), the lowest biaxial flexural strength values were recorded in the CopraPretty material.

Conclusion: Despite aggressive aging conditions, the materials showed high flexural strength values that could withstand intraoral forces. Therefore, clinical use of the materials is anticipated to be possible.

Öz

Amaç: Bu çalışmanın amacı, üç farklı yittria ile stabilize edilmiş tetragonal (t) zirkonya polikristalinin yaşlanma özelliklerini ve ayrıca düşük ısı bozunmasının materyallerin eğilme dayanımına etkisini incelemekti.

Gereç ve Yöntemler: Üç farklı markadan (Supra Zr, Zirkonzahn Prettau ve CopraPretty) 120 adet disk şeklinde örnek elde edildi. Her marka için kontrol gruplarındaki örnekler 5 saat boyunca 37 °C'de distile suda bekletildi. Deney gruplarındaki örnekler, yapay olarak üç farklı koşulda yaşlandırıldı. Örnekler termal döngü grubunda 5 °C ile 55 °C arasında 10.000 döngüye kadar yoruldu; Asetik Asit gruplarında %4 asetik asitte 80 °C'de 168 saat bekletildi ve otoklav yaşlandırma gruplarında 134 °C, 0,2 MPa'da 5 saat yaşlandırıldı. T monoklinik faz dönüşümü (Xm), tüm örneklerde X-ışını kırınımı kullanılarak ölçüldü. Her alt gruptan bir örnek yüzeyi taramalı elektron mikroskobu ile incelendi. Örneklerin eğilme dayanımı değerleri, tüm örnekler için çift yönlü eğilme dayanımı testi ile ölçüldü. Veriler istatistiksel olarak analiz edildi.

Keywords
Flexural strength, low-thermal degradation, monolithic zirconia

Anahtar Kelimeler
Eğilme dayanımı, düşük ısı bozunması, monolitik zirkonya

Received/Geliş Tarihi : 18.05.2022
Accepted/Kabul Tarihi : 22.06.2022

doi:10.4274/meandros.galenos.2022.91069

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Bulgular: En yüksek göreceli Xm miktarı otoklavda yaşlandırma gruplarında elde edildi (Supra Zr 0,36±0,01; Zirkonzahn Prettau 0,41±0,02; CopraPretty 0,58±0,04). İstatistiksel olarak Zirconzahn Prettau ve Supra Zr materyalleri arasında anlamlı bir fark yoktu (p>0,05), en düşük eğilme dayanımı değerleri CopraPretty materyalinde kaydedildi.
Sonuç: Agresif yaşlandırma koşullarına rağmen, materyaller ağız içi kuvvetlere dayanabilecek yüksek eğilme dayanımı değerleri göstermiştir. Bu nedenle materyallerin klinik kullanımlarının mümkün olması beklenmektedir.

Introduction

Monolithic zirconia ceramics are now recognized as promising materials with increased aesthetic and mechanical qualities in dental restorative processes (1).

Polymorphic zirconia crystals can exist in 3 different phases: monoclinic (m), tetragonal (t) and cubic. ZrO_2 can be mixed with other metallic oxides to achieve high molecular stability (2). Stabilized zirconia contains a metastable tetragonal phase that enables the usage of zirconium in bulk at room temperature. When material is subjected to mechanical stress, the metastable t phase which is resistant to crack propagation transforms into the m phase. Phase transformation from volume increases in localized areas around microcracks creates compressive stresses which prevents crack propagation - a phenomenon referred as transformation toughening (3). The metastable t phase transforms into the m phase starting from the material surface and into the bulk in a humid environment without mechanical stress. This causes microcracks and reduces the material strength. This process is called low-thermal degradation (LTD) or aging (4).

Despite its superior mechanical properties, zirconia is susceptible to aging, phase transformation in the presence of moisture, and undergoes LTD (5). Further knowledge is needed on the susceptibility of zirconia to LTD in order to increase clinical success (6).

Monolithic zirconia material must be addressed since the strength of dental restorative materials is an important foundation for appropriate restoration. This study centered on the impact of LTD on the biaxial flexural strength (BFS) of 3 monolithic yttria-stabilized tetragonal zirconia polycrystal (Y-TZP) ceramics and crystallographic phase changes of the materials' surfaces by analyzing X-ray diffraction, and surface characteristics by scanning electron microscope observation.

Consequently, the null hypotheses of the present study were that no significant difference among the monolithic zirconia materials and aging conditions have had no effect on the BFS of the materials.

Materials and Methods

Ethics committee approval for the study was obtained from the meeting of Selçuk University Faculty of Dentistry, Non-Interventional Clinical Research Evaluation Commission (protocol no: 2014/03; date: 25.03.2014).

Specimen Preparation

120 disc-shaped (12 mm Ø and 1.2±0.2 mm thickness) specimens were fabricated (Yenamak D40 milling machine, Yenadent Ltd., İstanbul, Turkey) from three different brands of monolithic zirconia: Supra Zr (Turkuaz Dental, İzmir, Turkey); Zirkonzahn Prettau (Zirkonzahn GmbH, Bruneck, Italy) and CopraPretty (Whitepeaks Dental, Wesel, Germany). At the pre-sintered stage, all specimens were wet-polished with silicon carbide papers (English Abrasives & Chemicals, London, UK) up to #1200 grit to achieve consistent smooth surfaces followed by the specimens were sintered according to the manufacturer's recommendations. Specimens were sintered at 1500 °C and 1600 °C (Supra Zr, 1500 °C; Prettau and CopraPretty, 1600 °C). Specimens were divided into four subgroups in each brand: Control, thermal cyclus, acetic acid ageing, and autoclave ageing. The specimens without any ageing conditions were designated as a control group. The test-groups designated are given in Table 1 together with the group abbreviations.

Artificial Aging

Control: Specimens were maintained in distilled water for five hours at 37°C in an incubator (EN 1200 model; Nuve, Ankara, Turkey).

Thermal cyclus: At thermal cycling machine (Nova, Konya, Turkey), specimens underwent 10.000 thermal

Table 1. Experimental groups with group abbreviations

	Condition			
Brands	Control	Thermal cyclus	Acetic acid ageing	Autoclave aging
Supra Zr	SC	ST	SA	SO
Prettau	PC	PT	PA	PO
CopraPretty	CC	CT	CA	CO

cycles between 5 and 55 °C with dwell time of 30 seconds and transfer time of 10 seconds.

Acetic acid aging: Specimens were immersed in 4% acetic acid solution (Puriss, Prolabo; Merck Group, Darmstadt, Germany) and kept in an incubator (EN 1200 model; Nuve, Ankara, Turkey) at 80±5°C for 168 hours.

Autoclave aging: Specimens were kept in a steam autoclave (DAC Professional Autoclave, Sirona Dental, NY, USA) at 134 °C, under two bars of pressure for five hours according to ISO 13356 standard.

X-ray Diffraction Analysis

The crystal structure was identified using an X-ray diffractometer (Bruker D8 Advance, Bruker AXS, Karlsruhe, Germany with monochromatic CuK radiation). The voltage and current were both set to 40 kV and 40 mA, respectively. With a step size of 0.0369, the specimen surfaces were scanned between 20 and 40 2Θ degrees (Lynxeye detector, Bruker AXS, Karlsruhe, Germany). Equation (1), based on the approach of Garvie and Nicholson (3), was used to compute the relative amount of m phase (Xm):

1) $$X_m = \frac{I_{m(111^-)} + I_{m(111)}}{I_{m(111^-)} + I_{m(111)} + I_{t(101)}}$$

$I_{t(101)}$ represents the intensity of a tetragonal peak whereas $I_{m(111\text{-})}$ and $I_{m(111)}$ reflect the intensities of m peaks.

Biaxial Flexural Strength Test

According to the ISO 687274 standard, the piston-on-three-balls method was used on a universal testing machine (ELISTA, TSTM02500, Elista Corp, İstanbul, Turkey) to estimate each specimen's BFS. Each specimen was put on three 3.2 mm diameter supporting steel balls that were equidistant from each other on a 10 mm diameter circle. A flat-end circular loading piston with a 1.4 mm diameter applied the load until the specimens fractured at a cross-head speed of 1 mm/min. The following formulae were used to calculate BFS (MPa) values [Equations (2-4)] according to the ISO 6872 standard:

2) $S = -0{,}2387\ P(X - Y)/d^2$

3) $X = (1+ v) \ln(r_2/r_3)^2 + [(1-v)/2]\ (r_2/r_3)^2$

4) $Y = (1+ v)\ [1 + \ln(r_1/r_3)^2] + (1-v)\ (r_1/r_3)^2$

where S is the BFS (MPa), P is the fracture load (N), v is the Poisson's ratio (0.25), r_1 is the support circle radius (mm), r_2 is the loaded area radius (mm), r_3 is the specimen radius (mm), and d is the specimen thickness at fracture origin (mm).

Scanning Electron Microscopy (SEM) Analysis

One specimen from each group was chosen randomly to reveal the surface topography by using SEM analysis. The specimen surfaces were cleaned in 96 per cent ethanol and allowed to air dry gently. The airborne-particle abraded surface of the specimens was coated with gold with a gold plating device (Sputter Coater 108 Auto, Cressington Scientific Instruments Ltd., Watford, England). The surfaces were examined at X1000 and X5000 magnifications (SEM-ZEİSS LS-10, England).

Statistical Analyses

The evaluation of the normal distribution and homogeneity of variances was done using the Kolmogorov-Smirnov and Levene tests, respectively. One-way ANOVA and Welch tests were used to analyze the data. Tukey HSD and Tamhane tests were used to detect the differences among subgroups. In order to clarify the relationship between the phase transformation and BFS, Pearson's correlation was used. The variability of the flexural strength was calculated using the Weibull distribution function. Each group's Weibull modulus (m) and characteristic strength (σ0) values were calculated. The Weibull modulus was calculated using the following formula (7):

5) $P_f(\sigma) = 1 - \exp[-(\sigma/\sigma_0)^m]$

where m is the Weibull modulus, Pf(σ) is the probability of failure, σ is the fracture strength, σ0 is the characteristic value that corresponds to 63.21 probability of failure, and. All statistical analyses were performed at a significance level of α=0.05.

RESULTS

Phase Transformation

The intensity of the tetragonal peak was recorded at 30°, whereas the intensities of m peaks were observed around 28 and 31 degrees, respectively.

The mean (± standard deviation) relative Xm of the subgroups are given in Table 2.

According to X-ray diffractometry (XRD) analysis, autoclave aging significantly affected the phase transformation in all groups. The highest amount of m phase transformation was detected in the CO group among the subgroups. There was no significant difference in phase transformation between SC and ST groups.

Biaxial Flexural Strength

Statistical results of BFS were shown in Table 3. The BFS values were affected by artificial aging. Control groups of Supra Zr and Prettau exhibited higher BFS values. Conversely, the BFS values of the CC, CT and CA groups were among the lowest of all the groups, and aging improved the BFS values of the autoclave and acetic acid groups of CopraPretty.

SEM Analysis

Representative SEM images are given in Figure 1. The results of the Xm values were consistent with the SEM images. When compared to the control groups, deep irregularities, pits, and grooves were observed on the surfaces of artificially aged groups. The CopraPretty material shows a different surface structure than other materials.

Weibull Analysis

Weibull analysis was used to analyze the variability of the BFS values of the materials. Characteristic strengths (σo), Weibull modulus (m), and R^2 values are given in Table 4.

According to the results of the Weibull analysis, it was observed that the m values of the materials differ according to the aging procedures. The low m value indicates the low reliability of these groups. The highest m value was found in the specimens in the SC group. This indicates that these specimens have similar BFS values and a low standard deviation, indicating that this material is more trustworthy.

Table 2. Mean (± SD) relative monoclinic phase (Xm) of subgroups (%)

	Supra Zr	Prettau	CopraPretty
Control	$0.28\pm0.03^{A,a}$	$0.29\pm0.02^{A,a}$	$0.11\pm0.01^{B,a}$
Thermal cyclus	$0.26\pm0.02^{A,a}$	$0.29\pm0.02^{B,a}$	$0.12\pm0.01^{C,a}$
Acetic acid	$0.29\pm0.03^{A,ab}$	$0.31\pm0.01^{B,ab}$	$0.18\pm0.02^{C,b}$
Autoclave aging	$0.36\pm0.01^{A,c}$	$0.41\pm0.02^{B,c}$	$0.58\pm0.04^{C,c}$

Subgroups that are not significantly different have the same superscript uppercase letters in the same column and the same superscript lowercase letters in the same row (Tukey's HSD and Tamhane tests, p>0.05)

DISCUSSION

This study purposed to elucidate the LTD properties of 3 types of monolithic Y-TZP ceramics by exposing specimens to various accelerated aging processes. To investigate the impact of accelerated aging treatment among materials the BFS, SEM analysis and XRD characteristics were examined. In light of the findings of the study, there was a significant difference among the monolithic zirconia materials and aging conditions have an effect on the BFS of the materials. As a result, the null hypotheses of the present study were rejected.

Three conditions were used for accelerated aging treatment to compare the effects of LTD in this study. Specimens in the control group were kept in distilled water in an incubator at 37 °C for 5 hours. The first condition was thermal cycling. Although the ISO TR 11450 standard (8) states that a thermo-cycling regimen comprising 500 cycles in water between 5 and 55 °C is a suitable artificial aging test, Gale and Darvell (9) came to the conclusion that 10,000 cycles equate to roughly one year of *in vivo* function. Hence the thermal cycling test used in this study was according to ISO TR 11450 standard (8) modified by Gale and Darvell (9). Acetic acid is the acid used for chemical stability testing, according to ISO standard 6872 (10). Kukiattrakoon et al. (11) used a modified method of ISO 6872 (10) in their study and immersed the specimens in 4% acetic acid at 80 °C for 168 h. The same condition was used for the acetic acid aging group in our study. Chevalier et al. (4) used a proportion of the m phase to compute the longevity of TZP and reported that 134 °C/5 h/0.2 MPa equates to 15 to 20 years in 37 °C water. The last condition was carried out in accordance with the ISO 13356

Table 3. Mean values ± standard deviations for biaxial flexural strength (MPa)

	Supra Zr	Prettau	CopraPretty
Control	$1178.116\pm35.022^{A,a}$	$1178.663\pm65.607^{A,a}$	$834.409\pm89.539^{B,a}$
Thermal cyclus	$1095.861\pm50.166^{A,b}$	$1109.320\pm112.012^{A,ab}$	$806.512\pm67.343^{B,a}$
Acetic acid	$984.863\pm168.067^{A,bc}$	$1045.040\pm61.091^{AB,b}$	$842.452\pm152.749^{AC,ab}$
Autoclave aging	$925.829\pm121.661^{A,c}$	$1014.606\pm107.040^{A,b}$	$963.102\pm96.573^{A,b}$

Same lowercase letters in the same column (Tukey's HSD, Tamhane's test; p>0.05) and same uppercase letters in the same row denote subgroups that were not significantly different

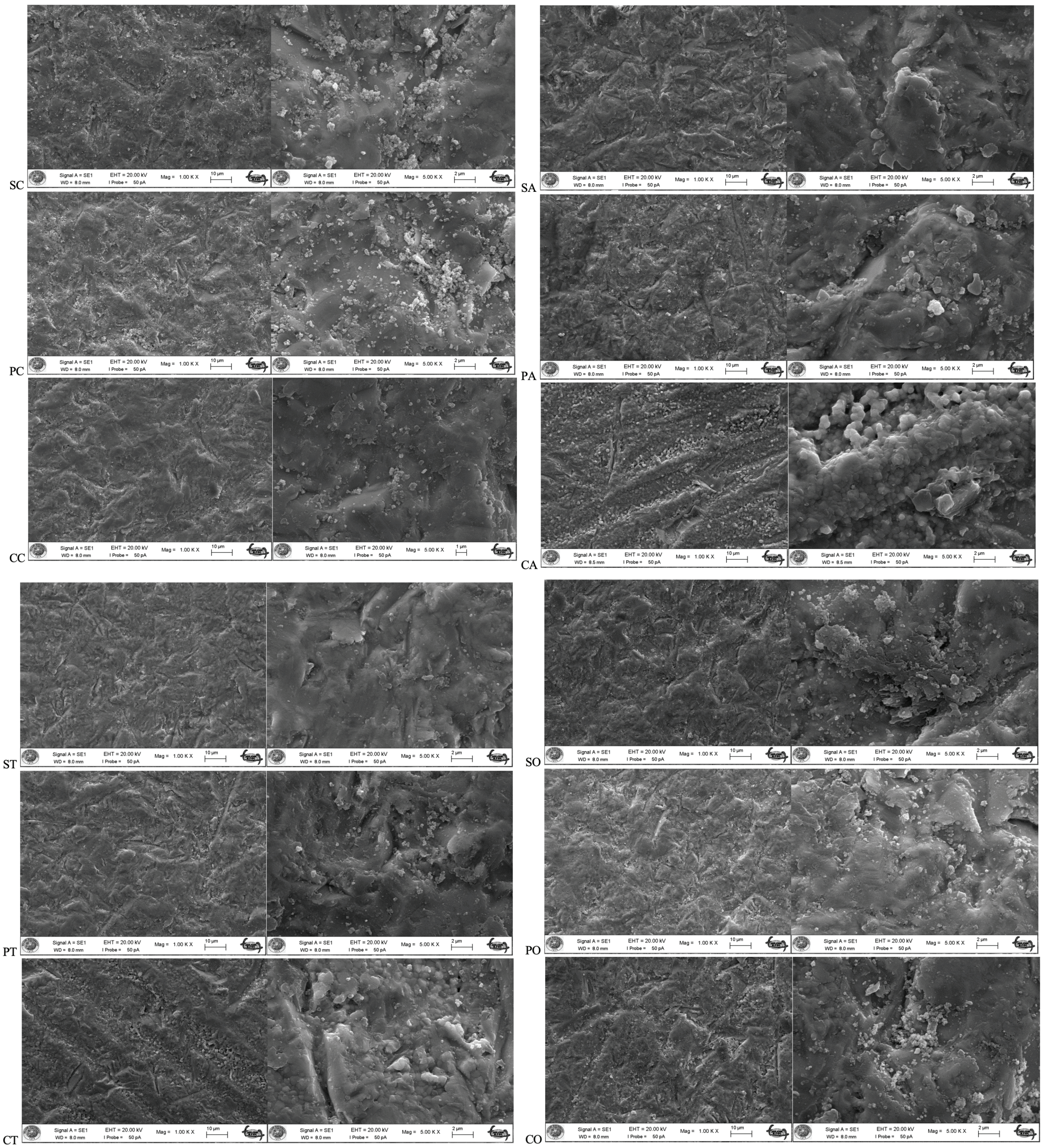

Figure 1. SEM images of experimental groups
SEM: Scanning electron misroscopy

(12) required accelerated aging test, as modified by Chevalier et al. (4).

The first conventional non-destructive technique to evaluate the zirconia transformation kinetics quantitatively was the XRD. Because the X-ray is several millimetres wide, it usually describes the behaviour of a section of the sample surface. However, the signal-to-noise ratio is quite low, available from

Table 4. Characteristic strengths (MPa), Weibull modulus (m) and R^2 values of groups

	Supra Zr			Prettau			CopraPretty		
	σ_o	m	R^2	σ_o	m	R^2	σ_o	m	R^2
Control	1196.00	35.62	0.734	1035.23	21.05	0.921	873.46	10.88	0.897
Thermal cyclus	1118.74	25.60	0.929	1159.75	11.22	0.896	836.62	13.91	0.854
Acetic acid	1054.24	6.72	0.913	1074.80	18.59	0.744	904.81	6.41	0.855
Autoclave aging	977.28	8.81	0.961	1061.29	10.93	0.973	1006.56	11.22	0.898

transformed regions where the converted zirconia fraction is less than 5% (13). XRD measurements have shown a sizable difference in m phase content according to aging conditions and zirconia materials in this study. The highest Xm values were found in the CO (0.58%±0.04) group while the lowest values were in CT (0.11%±0.01).

The biaxial flexural test which is the international standard for dental ceramics (14) was used in this study. The mean BFS values of the materials were 1046,16 MPa (Supra Zr), 1086,90 MPa (Prettau) and 861,61 MPa (CopraPretty) respectively. The BFS values of the control and thermal cycle groups of Supra Zr and Prettau materials were found to be higher than the acetic acid and autoclave aging groups. These results were consistent with a study done by Flinn et al., (15) which found that the flexural strength of the specimens decreased following accelerated aging treatment. However, the highest BFS values were obtained in the acetic acid and autoclave groups in CopraPretty material. These results support those of Pereira et al., (16) who found that volume expansion owing to phase transformation from t to m increase compressive residual stress and improves strength.

There was a negative correlation between phase transformation and BFS in Supra Zr and Prettau materials while a positive correlation occurred in CopraPretty material. While the highest Xm and the lowest BFS values were observed in the acetic acid and autoclave aging groups of Supra Zr and Prettau materials, both Xm and BFS values of acetic acid and autoclave aging groups of CopraPretty material were higher than the material's thermal cyclus and control groups. These results for CopraPretty material may be related to the transformation toughening phenomenon. Even though chemically similar materials were chosen, this apparent significant difference in sensitivity to LTD amongst identical ceramics could be ascribed to differences in manufacturing conditions, microstructure, and flaw distribution. This was also supported by the fact that changes in the surface layers of the materials were observed after accelerated aging treatment by SEM observation.

Two often used statistical measures to describe one aspect of structural reliability are the Weibull modulus (m) and characteristic strength. Assuming equivalent levels of mean strength between ceramics, higher Weibull modulus values correspond to materials with stronger structural reliability. The majority of ceramics are reported to have m values in the range of 5-15 (17). When compared to the other groups in the current study, the SC group displayed a higher Weibull modulus, demonstrating the material's higher clinical dependability. However, according to the m values, the materials have a tolerably low failure probability up to a bending stress level of around 500 MPa, which is higher than the stress caused by occlusal loads routinely measured intraorally (18).

Conclusion

Both hydrolysis and compressive residual stress can impact phase transformation to the m phase, and the major cause cannot be determined with certainty. The study was carried out with disc-shaped specimens in *in-vitro* conditions. Within the limitations of the present study in-vivo conditions with crown-shaped specimens may produce different results.

Acknowledgement

This article is extracted from my PhD dissertation entitled "Low Thermal Degradation of Three Yttria-stabilized Tetragonal Zirconia Polycrystalline and Effect of Low Thermal Degradation on Flexural Strength", supervised by Prof. Dr İsa Yöndem (Selçuk University, Konya/Turkey, 2015).

Ethics

Ethics Committee Approval: Ethics committee approval for the study was obtained from the meeting of Selçuk University Faculty of Dentistry, Non-Interventional Clinical Research Evaluation Commission (protocol no: 2014/03; date: 25.03.2014).

Informed Consent: Informed consent is not required.

Peer-review: Externally peer-reviewed.

Authorship Contributions

Concept: E.T.Ç., İ.Y., Design: E.T.Ç., İ.Y., Data Collection or Processing: E.T.Ç., İ.Y., Analysis or Interpretation: E.T.Ç., İ.Y., Literature Search: E.T.Ç., İ.Y., Writing: E.T.Ç., İ.Y.

Conflict of Interest: No conflict of interest was declared by the authors.

Financial Disclosure: This scientific research was supported by the Scientific Research Project Council of Selçuk University, Turkey (project no. 14102028).

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