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Effect of Sintering Holding Time on Tetragonal Phase Stability of Yttria Stabilized Zirconia Ceramics

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Abstract. In this study, the effect of second step sintering holding time on LTD of 3 mol% yttria tetragonal zirconia polycrystals (3Y-TZP) ceramic was studied. The Zirconia powder was die pressed followed by cold isostatically pressed (CIP) at 200 MPa, and the samples were initially heated to a first step temperature of 1400°C with constant heating rate 10°C/min and the samples were stand still for 1 minute. Then cooled down to the second step temperature of 1200°C and maintained at that temperature for 2 and 10 hours before cooled down to the room temperature. The other sample was sintered by using Single Step Sintering (SSS) at 1400°C for two hours. The phase content in the zirconia samples was measured by using X-ray diffractometer. Average grain size was calculated by using Field Emission Scanning Electron Microscope (FESEM). The low temperature degradation study was conducted in an autoclave containing superheated steam at 180°C and 10 bar pressures for periods up to 60 hours. Results revealed that the two-step sintering (TSS) effectively controlled the grain growth than SSS and subsequently ageing. Increase in second step sintering holding time from two to ten hours also increased the densification and hydrothermal ageing resistance of Y-TZP ceramics.

Introduction

Yttria stabilized tetragonal zirconia polycrystals (Y-TZP) are increasingly used as dental restorative material in recent years because they possess excellent mechanical properties, such as superior strength and fracture toughness while they also exhibit biocompatibility and aesthetic potential [1-3].The high strength and toughness of zirconia ceramics are due to stress-induced phase transformation toughening. Around a propagating crack, the metastable tetragonal (t) grains can transform to the monoclinic (m) structure, t–m transformation, and the associated volume expansion induces compressive stresses and eventually reduces or inhibits further crack propagation [2,4].However, the tetragonal grains can also spontaneously transform to monoclinic in a humid environment in the 20–300°C range [5,6] like in the oral cavity, a phenomenon being referred to as low temperature degradation (LTD) [5]. The mechanism of water incorporating into zirconia to induce t–m transformation is not yet fully understood, but it is well established that the degradation starts from the surface and proceeds inwards. Surface uplift [7, 8] and micro-cracks [9] are subsequently induced which can result in macrocracks [9] and surface roughening [10]. As a consequence, the mechanical [10,11] and even the aesthetic properties [12] of Y-TZPs are affected. The LTD susceptibility of Y-TZP is affected by various parameters. The degradation can be retarded by increasing the Y_2O_3 content. The sintering condition is a predominant factor influencing both the mechanical properties and LTD behaviour of 3Y-TZPs.However, the mechanism was not well studied and different manufacturers produce dental 3Y-TZP materials at different sintering conditions.

Two-step sintering (TSS) with a high first step sintering temperature and a lower second step holding temperature is a promising method used to obtain high-density bodies and smaller grain sizes [13-15]. In this work, the effect of TSS and second step sintering holding times on ageing and densification of 3Y-TZP ceramics was investigated.

Experimental Procedures

3 mol % yttria stabilizer tetragonal zirconia Powder (Kyoritsu Corporation, Japan) was used for green compact preparation. Green compacts were die pressed into discs (20 mm diameter). Before every pressing, the die and the punch wall were thoroughly cleaned with WD 40 lubricant. 2.5 g of powder was used for every discs. The pellets were slowly ejected from the die and used for further processing. Samples were sintered using a two-step sintering cycle with different holding times as reported in Sutharsini et. al. [15]. The sample was initially heated to 1400°C at a heating rate of 10°C/min and then maintained at that temperature for 1 minute. Thereafter the sample was cooled down to 1200°C (holding temperature) at the same cooling rate (10°C/min) and maintained at that temperature for varying holding times of 2 and 10 hours, before cooled down to the room temperature [13]. Table 1 shows the designations of the samples used with corresponding sintering profiles.

Table 1. Sample designations.

Designation	1st step	2nd step
SSS	1400°C at 2 hrs.	
TSS2	1400°C at 1 min.	1200°C at 2 hrs.
TSS10	1400°C at 1 min.	1200°C at 10 hrs.

Phase analysis of the samples was carried out by X-ray diffraction (XRD) (EMPYREAN, PAN analytical, Netherlands) with Cu-K_α radiation source at room temperature. Average grain size was calculated by using Field Emission Scanning Electron Microscope images (FESEM, Carl Zeiss Auriga). The samples were polished and thermally etched before imaging. The hydrothermal ageing/low temperature degradation study was conducted in a sealed autoclave, containing superheated water steam at 180°C and 10 bar pressure for periods up to 60 hours.

Results and Discussion

The density of the sintered samples was measured by water immersion technique, and the relative density was calculated by considering the theoretical density of 3Y-TZP as 6.09 gcm^{-3}. The SSS sample showed maximum density of 98.4 %. The relative density of TSS2 and TSS10 samples were 95.8 % and 97.2 %. It can be understood that the lower relative densities of TSS2 and TSS10 samples due to lower sintering time at 1400°C. However, the relatively density was increased with increasing holding time which indicates that the densification occurred during the second step sintering. Fig. 1 (a), (b) and (c) illustrates the FESEM images of SSS, TSS2 and TSS10 samples. From the images, we calculated the average grain sizes. The average grain size of SSS sample was found as 450 (±39) nm and the average grain size of TSS samples were found as 230 (±22) and 234 (±19) nm for 2 and 10 hours holding time respectively. From the grain size measurements, we can conclude that the TSS suppressed the grain growth. Also, in TSS samples, grain growth in 3Y-TZP ceramics was not depend on holding time [15].

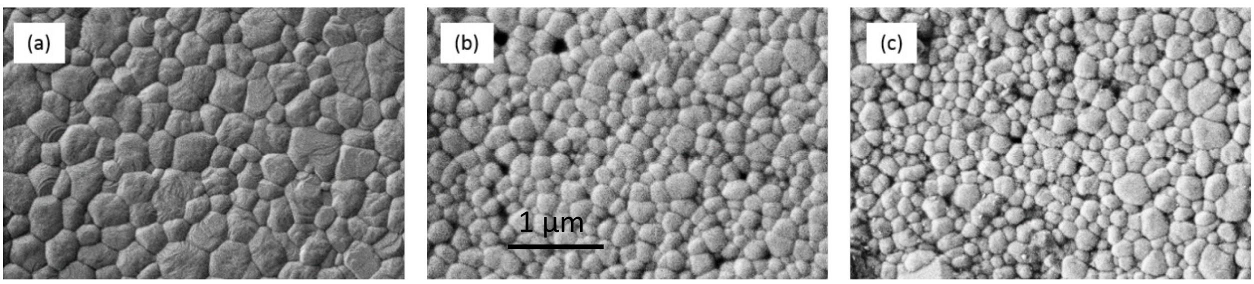

Fig. 1. FESEM images of (a) SSS, (b) TSS2 and (c) TSS10 samples.

According to the XRD analysis, as sintered samples using 2 and 10 hours holding time and SSS samples exhibited 100% tetragonal phase. The ageing studies was conducted all samples up to 60 hrs. Fig. 2 shows the XRD spectrum of samples aged for 60 hours. Table 2 summarizes the monoclinic phase content of the samples with ageing time. SSS sample showed 5 % tetragonal to monoclinic phase transformation after 2 hours of ageing and it increased up to 90 % after 60 hours of ageing. On the other hand, there were no tetragonal to monoclinic transformation observed in both TSS samples up to 24 hours of ageing. The TSS2 sample showed 4 % of monoclinic phase content after 60 hours of ageing and TSS10 did not show any phase transformation.

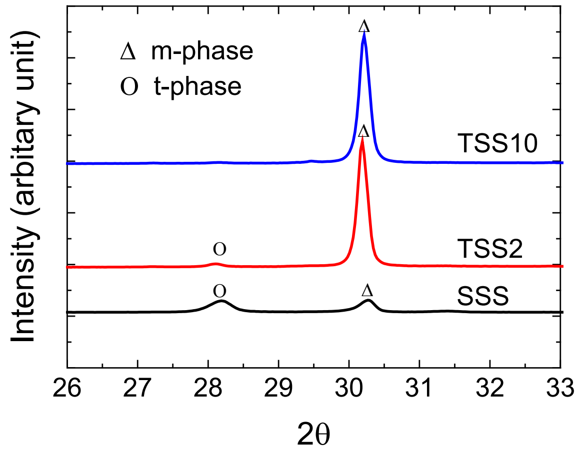

Fig. 2. XRD spectrum for 3Y-TZP samples sintered using SSS, TSS2 and TSS10 sintering profiles after 60 hours of ageing.

Table 2. Percentage of monoclinic content in the samples after different ageing time.

Sample	**Monoclinic % after ageing time**				
	2 hours	6 hours	12 hours	24 hours	60 hours
SSS	5 %	75 %	48 %	87 %	90 %
TSS2	0 %	0 %	0 %	0 %	4 %
TSS10	0 %	0 %	0 %	0 %	0 %

The ageing studies revealed that the TSS samples show better ageing resistance against SSS sample. Especially, the sample sintered with 10 hour holding time showed better ageing resistance against all other samples. It has been reported that the grain size plays a vital role in ageing [14,16-18]. With this we can explain the poor ageing resistance of SSS sample which has the larger grain size among all samples. On the other hand, there were no significant changes observed in the average grain size of TSS2 and TSS10 samples. However, the better ageing resistance of TSS10 sample with same grain size open more questions on the role of grain size in ageing. Few studies were reported that the surface yttria content and structural defects also play vital role in ageing. However, more works needed on concluding the ageing mechanism and other determining factors of ageing in 3Y-TZP ceramics.

Conclusion

We successfully controlled the grain growth of 3 mol% yttria tetragonal zirconia polycrystals and subsequently controlled the ageing by applying two step sintering. The densification was found comparatively lower in two step sintered samples. The grain growth was controlled, and densification was increased with increasing second step sintering holding time. Tetragonal to monoclinic phase transformation was found to increase with increasing ageing time. The sample fabricated with 10 hours holding time exhibited excellent ageing resistance even after 60 hours of ageing.

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