Effects of the sintering conditions of dental zirconia ceramics on the grain size and translucency

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PURPOSE. This study aimed to identify the effects of the sintering conditions of dental zirconia on the grain size and translucency. **MATERIALS AND METHODS.** Ten specimens of each of two commercial brands of zirconia (Lava and KaVo) were made and sintered under five different conditions. Microwave sintering (MS) and conventional sintering (CS) methods were used to fabricate zirconia specimens. The dwelling time was 20 minutes for MS and 20 minutes, 2, 10, and 40 hours for CS. The density and the grain size of the sintered zirconia blocks were measured. Total transmission measurements were taken using a spectrophotometer. Two-way analysis of variance model was used for the analysis and performed at a type-one error rate of 0.05. **RESULTS.** There was no significant difference in density between brands and sintering conditions. The mean grain size increased according to sintering conditions as follows: MS-20 min, CS-20 min, CS-2 hr, CS-10 hr, and CS-40 hr for both brands. The mean grain size ranged from 347-1,512 nm for Lava and 373-1,481 nm for KaVo. The mean light transmittance values of Lava and KaVo were 28.39-34.48% and 28.09-30.50%, respectively. **CONCLUSION.** Different sintering conditions resulted in differences in grain size and light transmittance. To obtain more translucent dental zirconia restorations, shorter sintering times should be considered. *[J Adv Prosthodont 2013;5:161-6]*

KEY WORDS: Grain size; Sintering; Translucency; Transmittance; Y-TZP; Zirconia

INTRODUCTION

All-ceramic restorations are preferred by both dentists and patients because of their good esthetic properties compared to metal restorations, namely an inherent translucency similar to that of natural teeth.[1] However, all-ceramic restorations have limited indications and are difficult to be applied to a challenging prosthesis that requires physical strength because of their intrinsic weaknesses.[2]

In recent years, 3 mol% stabilized tetragonal zirconia polycrystalline (3Y-TZP) ceramics have gained favor as restorative materials because of their excellent mechanical properties[3-5]; they have high strength and toughness, good biocompatibility,[6-8] and relatively good esthetic properties.[3]

Esthetically fabricated all-ceramic prostheses should have a color and translucency comparable to those of natural teeth.[9,10] Of these two factors, the translucency of the core is the most important determinant of the esthetic

properties of all-ceramic restorations.[1] However, the zirconia core is less translucent than other dental all-ceramic materials such as glass-infiltrated ceramic, which is known for its good esthetic properties.[11-13] Therefore, by increasing the translucency of the zirconia core, the esthetic characteristics of dental restoration can be improved.

Fundamentally, translucency is highly dependent on light scattering.[14] Pores inside materials greatly influence light scattering; low porosity is required to improve the translucency of ceramics.[15,16] In addition, the amount of light absorbed, reflected, and transmitted depends on the amount of crystal and chemical components inside the core materials, the size of the particles compared with the wavelength of incident light.[17]

Translucency of zirconia is related to the amount and type of additives,[18] the sintering temperature,[19] the atmospheric conditions during the sintering process,[20] and the heating methods.[21] In particular, the final temperature of the sintering process and the heating method used are direct determinants of the density, porosity, and grain size of zirconia.[22]

Several studies have reported that small mean grain size is associated with enhanced translucency, mechanical properties and a delay in low temperature degradation (LTD) caused by inhibition of the transformation from the tetragonal phase to the monoclinic phase.[18,21,23] Microwave sintering (MS) is more efficient than conventional sintering (CS) at reducing the size of zirconia grains.[22] MS has advantages of improved productivity and energy saving due to volumetric heating and rapid sintering over CS, and yields zirconia grains with a uniform microstructure.[24,25] For ceramics, improved densification and inhibition of the grain growth may reduce the sintering time by enhancing the mechanical properties and enabling sintering at lower temperatures.[18,26] However, the translucency of dental zirconia in relation to MS has not been extensively studied.

The aim of this study was to investigate the pattern of changes in the density, grain size, and translucency of dental zirconia according to various sintering conditions (time and method), as well as the effects of the sintering conditions on the translucency of dental zirconia. Our hypothesis in this study was that a change in the sintering conditions would change the translucency of the zirconia core.

MATERIALS AND METHODS

Commercially available Lava frame zirconia (3M ESPE, St. Paul, MN, USA) and KaVo Everest ZS-blanks (KaVo Dental GmbH, Biberach, Germany) dental zirconia blocks were used in this study. The sintering temperatures for MS and CS were set according to the instructions of the manufacturers. The heating rate was 50℃/min for MS and near 10℃/min for CS. The dwelling time was 20 minutes for MS and 20 min 2, 10, or 40 hr for CS (four conditions). The detailed sintering conditions are shown in Table 1. After sintering, 10 specimens [10 mm (width) × 10 mm (length) × 1 mm (thickness)] were made for each of the two types of zirconia (Lava and KaVo) and for each of the five sintering conditions using a diamond saw (ISOMET, Buehler Ltd., Lake Bluff, IL, USA). The blocks were cut in a container filled with water using low speed diamond saw. The thickness of the core was adjusted to 1 mm (± 0.05 mm) using electronic Vernier calipers (Mitutoyo, Kanagawa, Japan). The core was then polished using 1,200-grit wet sandpaper (Buehler Ltd., Lake Bluff, IL, USA).

The density of the sintered specimens was measured using the Archimedes method in water, following the ISO 18754 standard.[27]

To measure the translucency of the dental zirconia cores, light transmittance was measured using a spectrophotometer. (SpectraMagic CM-3500d, KONICA MINOLTA, Osaka, Japan). Measurements were performed at wave-

Table 1. Sintering conditions used for the various zirconia specimens

Brand	Group (n=10)	Sintering Temperature (℃)	Heating rate (℃/min)	Rising time (min)	Dwell time (hr)
Lava	MS – 20 min	1,500	50	30	0.33
	CS – 20 min	1,500	10	150	0.33
	CS – 2 hr				2
	CS – 10 hr				10
	CS – 40 hr				40
KaVo	MS – 20 min	1,450	50	30	0.33
	CS – 20 min	1,450	7	210	0.33
	CS – 2 hr				2
	CS – 10 hr				10
	CS – 40 hr				40

lengths ranging from 400 to 700 nm at 10 nm intervals. Measurements were repeated three times per specimen.

Transmittance (T) was determined using the following equation:

$$T = (L\ specimen\ /\ L\ source) \times 100\%,$$

where L specimen is the specimen luminance and L source is the source luminance.

To measure the size and microstructure of the grain, the blocks were polished using a 1 μm diamond paste. All specimens were thermally etched at 1,400℃ for 10 min. Afterwards, all specimens were cleaned ultrasonically in acetone for 10 min and then dried for 24 hr. The specimens were then investigated via scanning electron microscopy (JSM-6360, JEOL Techniques, Tokyo, Japan). The grain size was calculated using the average linear intercept method of ASTM Standard E112.[28] For each specimen, 25 line segments were taken into account.[29] This process was performed last to prevent interference from the effects of the polishing that was done to obtain SEM images to measure light transmittance.

Descriptive statistics were expressed as mean and standard deviation (SD). To analysis relationship with 2 factors, brands and sintering methods, the 2-way analysis of variance (ANOVA) model was used to compare the density, grain size, and light transmittance values (%) of Y-TZP according to different sintering conditions and zirconia brands. As assumption of normality tested by the Shapiro-Wilk's test was satisfied in the distribution of density and grain size, the (parametric) 2-way ANOVA was applied. For density all 2 factors were insignificant. For grain size, the interaction term was insignificant in the full factorial model, therefore the main-effect model was used. However the assumption of normality was not satisfied in that of light transmittance and nonparametric version of 2-way ANOVA using rank transformed value was applied. The interaction term was significant with full factorial model of light transmittance. As assumption of equal variance was not satisfied in grain size and transmittance, the Tamhane's T2 method was applied for the post-hoc comparison. All statistical tests were performed at a type-one error rate of 0.05. The statistical software package SPSS version 12.0 (SPSS V12.0K, SPSS Inc., Chicago, IL, USA) was used.

RESULTS

The density of the Lava zirconia core ranged from 6.05 to 6.07, and that of the KaVo zirconia core from 6.05 to 6.06. The density of Lava was highest (6.07) at MS-20 minutes and the lowest at CS-40 hours, whereas the density of KaVo was highest at both MS-20 minutes and CS-2 hours. Under all conditions, the density of each specimen was close to the full density. The density of Lava did not differ significantly from that of KaVo. Furthermore, there was no significant difference in density according to sintering conditions.

The changes in the mean light transmittance according to the brands and sintering conditions are summarized in Table 2. The mean light transmittance values of Lava and KaVo were 28.39-34.48% and 28.09-30.50%, respectively. Both Lava and KaVo showed the highest mean light transmittance values of 34.48% and 30.50%, respectively, at MS-20 minutes, and the lowest mean light transmittance values of 28.39% and 28.09%, respectively, at CS-40 hours. Statistical analyses revealed that sintering conditions interacted with brand.

The mean grain size ranged from 347-1,512 nm for Lava and 373-1, 481 nm for KaVo according to the sintering conditions. Both Lava and KaVo had the smallest mean grain sizes of 347 and 373 nm, respectively, at MS-20 minutes, and the largest mean grain sizes of 1,512 and 1,481 nm, respectively, at CS-40 hours (Table 3). The longer the sintering time, the larger the resulting grain. This finding was confirmed by SEM images (Fig. 1).

Table 2. Comparative light transmittance (%) according to two zirconia brands and five sintering conditions (n=10)

	Lava	KaVo
MS – 20 min	34.48 (0.24)[a]*	30.50 (0.37)[b]
CS – 20 min	30.32 (0.64)[b,c]	29.62 (0.20)[d]
CS – 2 hr	29.80 (0.32)[c,d]	28.61 (0.31)[e,f]
CS – 10 hr	28.86 (0.16)[e]	28.39 (0.43)[f,g]
CS – 40 hr	28.39 (0.19)[f,g]	28.09 (0.37)[g]

*Different superscripts indicate significantly different group (*P*<.05).

Table 3. Comparative grain size (nm) according to two zirconia brands and five sintering conditions (n=25)

	Lava	KaVo
MS – 20 min	347 (26)[A,a]*	373 (18)[B,a]
CS – 20 min	477 (17)[A,b]	415 (45)[B,b]
CS – 2 hr	603 (40)[A,b]	605 (70)[B,b]
CS – 10 hr	735 (84)[A,c]	912 (58)[B,c]
CS – 40 hr	1512 (172)[A,d]	1481 (149)[B,d]

*Different superscripts indicate significantly different group (*P*<.05): upper cases for brands and lower cases for sintering conditions.
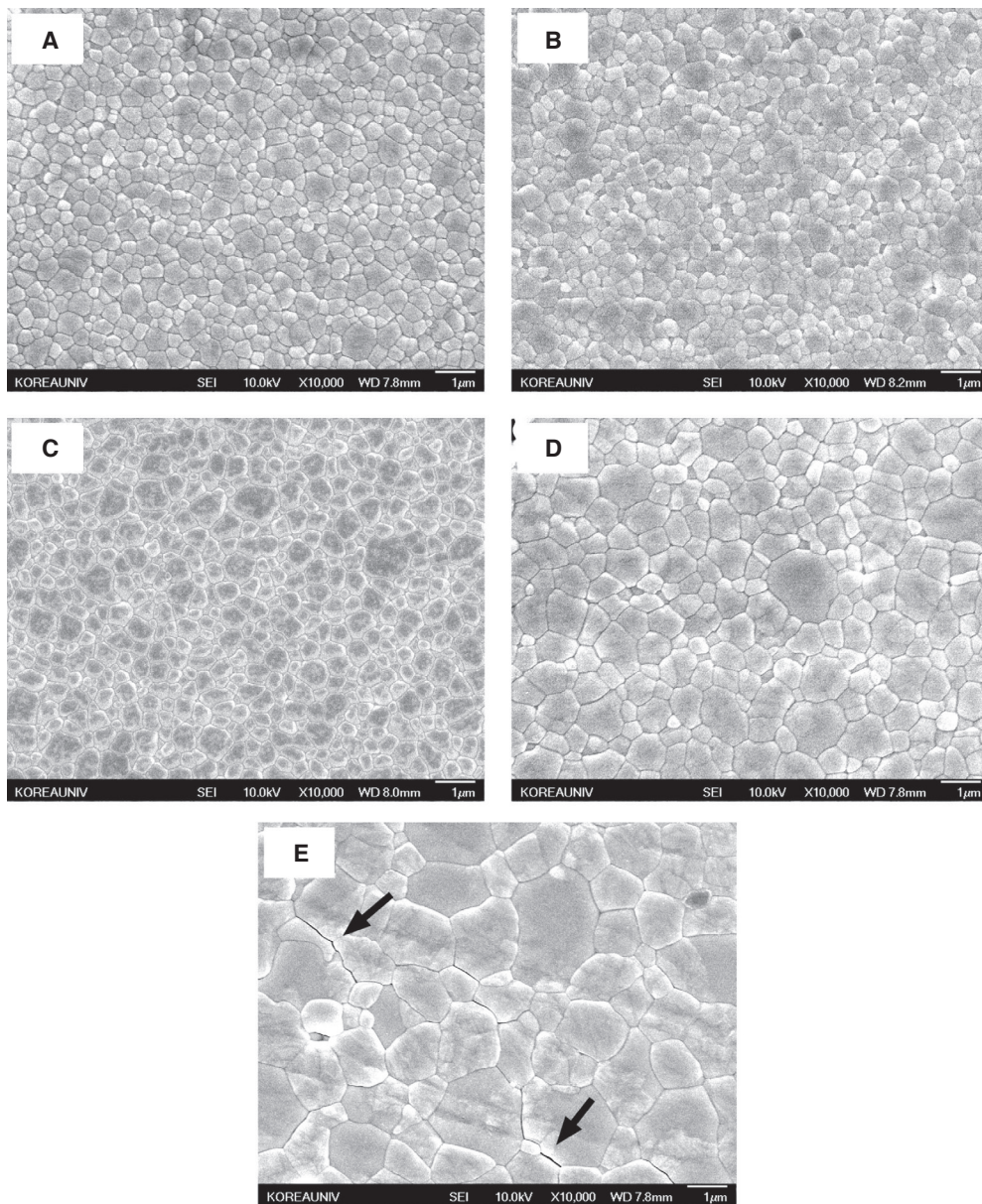
Fig. 1. SEM images (Original magnification ×10,000) of the polished and thermally etched surface of Lava specimens sintered for various times using different approaches: A: MS-20 minutes, B: CS-20 minutes, C: CS-2 hours, D: CS-10 hours, and E: CS-40 hours. The black arrows indicate grain boundary cracks.

DISCUSSION

Because we found that the grain size ($P<.001$) and translucency ($P<.001$) of dental zirconia changed according to changes in sintering conditions, we adopted our null hypothesis.

The translucency of dental ceramics can be studied in three ways: through direct transmission, which can be assessed by measuring the light that reaches a detector; total transmission, which can be assessed by measuring both the light that reaches the detector and that which passes the ceramic and is scattered; and indirect measurements via spectral reflectance.[14] In this study, we measured translucency by assessing the total transmission.

Light scattering has been widely studied owing to its important relation to the esthetics of all-ceramic restorations affected by color and translucency.[14,30] Light scattering relies on the grain size and wavelength of incident light. If the grain and wavelength of incident light are in a similar range, the amount of light scattering increases with grain size and lowers the light transmittance. If grain size is much larger than the wavelength of incident light, the amount of

light scattering becomes inversely proportional to the grain size, and independent of the wavelength of incident light.[18,31,32]

In our study, light transmittance decreased with an increase in grain size not only for MS and CS, which have grain sizes in the range of visible light, but also for the larger-grained CS10 and 40. Our results are consistent with Hayashi et al.[33] in which an increase in grain size led to higher light transmittance when experimenting with a wavelength of 300-1,700 nm and 0.82-1.43 μm particles. Also, O et al.[34] reported that the measurement of light transmittance with a grain size of 0.8-2 μm using a 1,000 nm wavelength, which is consistent with our results.

Thus far, factors having a negative effect on translucency are known to be grain boundaries, pores, second-phase of component, and light scattering from rough surfaces.[33] However, recent studies reported that polycrystalline materials have more light scattering in micropores than at grain boundaries despite their birefringence property.[33,35,36] Even so, when grain size falls below micron units, the effect of light scattering from pores within materials is significantly reduced.[18,20,33,35]

The theoretical full density of the TZP material was 6.10 g/cm^3, and ISO 13356 states that it is appropriate for the density of zirconia to exceed 6.00 g/cm^3.[37] In this study, the density of all the specimens ranged from 6.06 to 6.07 g/cm^3. There were no statistically significant differences among specimens. Hayashi et al.[33] reported no relationship between pores and light scattering for specimens having a submicron-sized grain and high density (> 99%), which is consistent with our results. The slight difference of final sintering temperatures caused by different manufacturer`s instructions (1,450 and 1,500℃) did not affect the densities of the zirconia specimens.

Tekeli and Erdogan[38] reported that a high sintering temperature and a long dwelling time increased the grain size and thus, the number of micropores, resulting in a material with reduced mechanical properties. In our study, CS resulted in a difference in grain sizes based on the dwelling time (P<.001). In particular, the products of both manufacturers showed minute grain boundary cracks at CS-40 hours (Fig. 1E). It has been reported that although a grain boundary crack is a type of defect that decreases light transmittance, the effect is very weak.[33,35,36] Therefore, this suggests that an excessive dwell time decreases the light transmittance mainly by inducing grain growth.

In contrast, Hjerppe et al.[39] reported that a short sintering time for dental zirconia decreased the grain size, though this finding was not statistically significant, and had no influence on the mechanical properties. In our study, however, there was a significant difference in the heating rates of MS and CS, which indicates a great difference in overall entire sintering time and mean grain size (Table 3).[16]

We found that a short sintering time yielded a small grain with high light transmittance while, in contrast, large grains formed by long sintering times had low light transmittance (Table 1 and 2). We found that the MS method, which can be completed in a short time, yielded zirconia with a small grain size and high light transmittance. These results should improve the low light transmittance of dental zirconia compared to other glass-infiltrated ceramics.[11,12] In addition, considering the clinical significance of zirconia, the physical properties and marginal fitness of the coping should be studied in relation to the sintering method used, grain size, and light transmittance.

CONCLUSION

Within the limitations of this study, we concluded that sintering condition (time) affected the grain size of dental zirconia: the shorter the sintering time, the smaller the grain size and this decrease of grain size resulted in an increase of the light transmittance values of the final dental zirconia.

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