Asian Journal of Scientific Research

ISSN 1992-1454

science alert
http://www.scialert.net

ANSInet
an open access publisher
http://ansinet.com

Asian Journal of Scientific Research 8 (3): 342-350, 2015
ISSN 1992-1454 / DOI: 10.3923/ajsr.2015.342.350

Development of Translucent Zirconia for Dental Crown Applications

[1]Kai Yuan Theng, [1]Andanastuti Muchtar, [2]Norziha Yahaya and [1]Mariyam Jameelah Ghazali

[1]Department of Mechanical and Materials Engineering, Faculty of Engineering and Built Environment, Universiti Kebangsaan Malaysia (UKM), 43600, Bangi, Selangor, Malaysia

[2]Department of Prosthodontics, Faculty of Dentistry, Universiti Kebangsaan Malaysia, 50300, Kuala Lumpur, Selangor, Malaysia

Corresponding Author: Andanastuti Muchtar, Department of Mechanical and Materials Engineering, Faculty of Engineering and Built Environment, Universiti Kebangsaan Malaysia (UKM), 43600, Bangi, Selangor, Malaysia Tel: +603-8921 3895

ABSTRACT

Zirconia-based dental ceramic is widely used for crown restoration, because of its superior mechanical properties and favorable biocompatibility. However, yttria-stabilized tetragonal zirconia (3YSZ), which is used in most dental crown restorations, has low translucency. This characteristic is unfavorable and results in low aesthetic quality of dental restoration. In this study, translucent 3YSZ dental ceramics were fabricated using nano-sized powder (20 nm) and high-temperature sintering (1500°C). The green bodies were slip casted and consolidated with cold iso-static pressing to create compact bodies. During the slip casting process different amounts of dispersing agent Polyethyleneimine (PEI) (0.2, 0.4, 0.6 and 0.8 wt%), were used to prevent agglomerates and create homogeneous suspensions. A minute amount (0, 0.1, 0.2, 0.3 and 0.4 wt%) of alumina was added into the suspension as sintering aid. The translucency or light transmittance of 3YSZ specimens was measured by an ultraviolet-visible spectrometer. Results showed that the 3YSZ specimen with 0.4 wt% alumina and 0.4 wt% PEI exhibited the highest light transmittance. The specimen also had larger grain size, because of excessive grain growth. The Vickers hardness of the specimens was insignificantly affected by the amount of alumina and PEI addition.

Key words: Dental ceramics, translucency, yttria-stabilized tetragonal zirconia

INTRODUCTION

Ceramics have been used in dental crown applications because of their biocompatibility and good aesthetic properties. Although, metal alloy restorations have good mechanical strength, metal alloy performs poorly in the aesthetic aspect because of its undesirable metallic color. Aesthetic properties play an important role in dental restorations, because the result is expected to match the natural appearance of a human tooth (Li *et al*., 2010; Chang, 2010; Spyropoulou *et al*., 2011). The factors that control the aesthetic quality of dental restorations include the restoration size, shape, color, texture and translucency of the material (Jiang *et al*., 2011). Among these criteria, core translucency is a critical factor that controls the aesthetic quality of dental ceramics.

Photon can be absorbed, transmitted or reflected when light strikes the surface of an object. Translucency or light transmittance is the physical property of a material that allows a fraction of incident light to pass through the medium. Translucency of a dental ceramic highly depends on its light-scattering property. Translucency could not be achieved, when the incident light is distributed by scattering centers in the microstructure, such as pores and grain boundaries

(Apetz and van Bruggen, 2003; Alaniz *et al*., 2009). However, light scattering is inhibited, when the scattering center is smaller than the wavelength of the incident light, resulting in translucency (or even transparency) of the material. Thus, controlling the scattering centers of a material is the key in developing a translucent material.

Three mole percent yttria-stabilized zirconia (3YSZ) is one of the materials widely used in dental restoration. The 3YSZ has been used to fabricate dental crowns, bridges and fix prostheses. The 3YSZ is bio-inert, exhibiting excellent mechanical strength, because of its transformation toughening. However, 3YSZ has low translucency, which makes satisfying the aesthetic requirement difficult for such material (Luo and Zhang, 2010). Therefore, controlling the chemical composition and scattering centers in the microstructure of 3YSZ is important to achieve good core translucency.

The microstructure of 3YSZ is greatly influenced by the size of the powder used during the consolidation process. The 3YSZ with smaller pores and grain size can be fabricated using micron or nano-sized powder. Grain size smaller than the wavelength of visible light (380-780 nm) inhibits light scattering in 3YSZ, improving the translucency of the material (Kim *et al*., 2013; Jiang *et al*., 2011) compared the light transmittance of 3YSZ fabricated with two different powder sizes, namely, 40 and 90 nm. They reported that 3YSZ specimen with 40 nm powder had better light transmittance and higher relative density at a sintering temperature of 1350°C. The specimen with 40 nm powder had better light resistance, because a smaller powder size has larger surface area, which helps decrease the activation energy needed during the sintering process. However, 3YSZ specimens can be sintered to nearly 99% of relative density at a sintering temperature of 1500°C regardless of the size of the powder used. Thus, the findings of Jiang *et al*. (2011) indicate that the full densification of 3YSZ occurs at a sintering temperature of 1500°C.

Grain size homogeneity is another way to control scattering centers in the 3YSZ microstructure. Grain size homogeneity leads to uniform density that can minimize the formation of pores throughout the 3YSZ microstructure. The consolidation technique used, when forming the green body is important to determine the grain size homogeneity of the sintered product (Huo *et al*., 2012; Mouzon *et al.,* 2008) compared two consolidation techniques, namely, uniaxial pressing and slip casting. They reported that slip casting produced a sample with better packing homogeneity. However, using slip casting as a consolidation technique has its drawback. The yttria suspensions age when used in the slip casting process, causing rapid growth in the suspension density leading to agglomeration (Mouzon *et al*., 2008). Agglomeration is undesired, because this phenomenon causes different density gradients in the green bodies of 3YSZ and ultimately promotes pores formation. Agglomeration can be avoided by introducing a dispersing agent into the 3YSZ slurry. Amat *et al*. (2014) studied the optimum amount of dispersing agent Polyethyleneimine (PEI) to be added to the 3YSZ suspension and reported that adding 0.5 wt% of PEI provides the lowest viscosity, indicating that the suspension had the lowest agglomeration.

Different additives have been studied and used as sintering aids to fabricate a 3YSZ with higher density and better microstructure homogeneity (Lopez-Honorato *et al*., 2012). Recent studies have suggested that adding a small amount of alumina could stabilize the tetragonal phase of 3YSZ and increase the densification of the sintered bodies (Chen *et al*., 2008). According to Chen *et al*. (2008), the limiting solubility of alumina in zirconia is approximately 0.2 wt%. However, the optimum amount of alumina addition into 3YSZ with PEI as dispersing agent and nano-sized zirconia powder has not yet been fully established. Therefore, this study investigates the effect of alumina and PEI addition on the translucency, density, hardness and microstructure of 3YSZ. The green body is fabricated by slip casting, with the maximum sintering temperature of 1500°C to achieve full densification.

MATERIAL AND METHODS

The nano-sized 3YSZ powder was supplied by Nabond Technologies (China). The 3YSZ powder has an average size of 20 nm and specific surface area of 50 m^2 g^{-1}. Figure 1 shows the morphological characterization of the 3YSZ powder in the transmission electron microscopy image. The dispersant agent PEI was supplied by Sigma-Aldrich and has an average molecular weight of 50,000. The alumina used was supplied by Hasrat Bestari Sdn. Bhd., with a particle size of 50 nm and specific surface area of 18 m^2 g^{-1}.

Preparation of 3YSZ green body: The green body of the 3YSZ specimens was fabricated by slip casting. The slip casting suspensions were prepared by mixing 20 wt% of 3YSZ powder with distilled water. Different proportions of alumina powder (0, 0.1, 0.2, 0.3 and 0.4 wt%) and PEI (0.2, 0.4, 0.6 and 0.8 wt%) were then added into the 3YSZ suspensions. Subsequently, the mixtures were stirred by a magnetic stirrer for 45 min and then submerged in an ultrasonic bath for 15 min to break any agglomeration that formed in the suspensions. The slip casting process of the 3YSZ suspension was conducted using a 25 mm diameter mold placed on plaster of Paris at room temperature. The suspensions were air-dried and the green body formed after 24 h.

Cold isostatic pressing and sintering of 3YSZ: Cold Isostatic Pressing (CIP) was used to increase the green body density. Cold isostatic pressing was performed at a pressure of 200 MPa and pressing time of 60 sec. After CIP, the green bodies were sintered at 1500°C in a furnace (CMTS Furnace L16) with a soaking time of 2 h. The sintering profile is shown in Fig. 2.

Sample characterization: The light transmittance of the fabricated samples was determined using an ultraviolet-visible spectrometer (Perkin Elmer Lambda 35). Light transmittance is obtained using the equation:

$$T = \frac{L}{L_0} \times 100\%$$

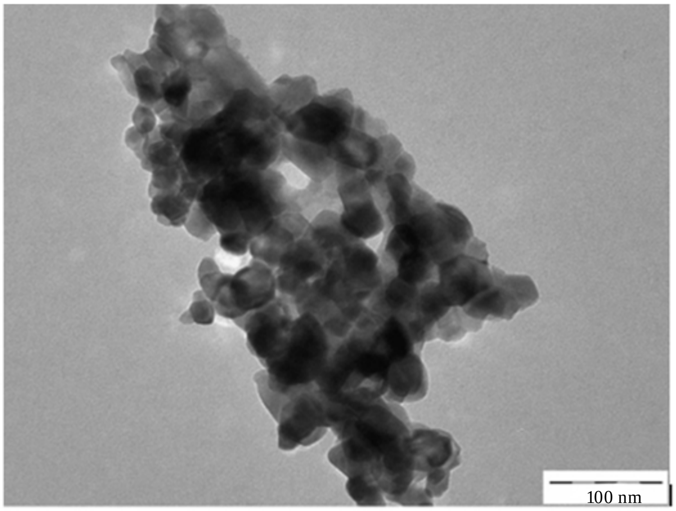

Fig. 1: TEM image of the 3YSZ powder

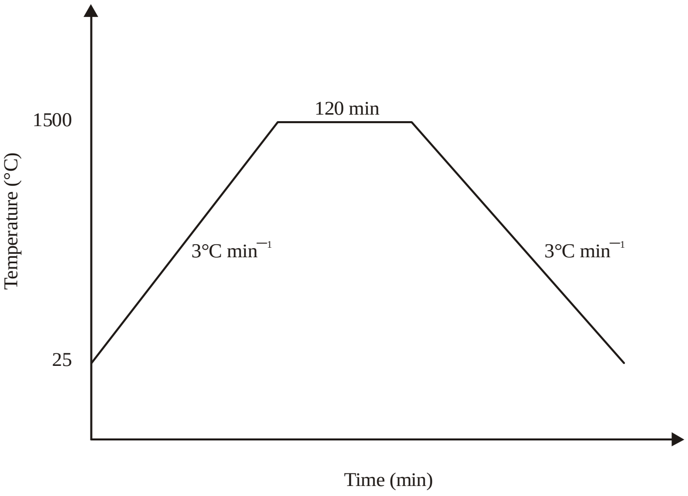

Fig. 2: Sintering profile of 3YSZ

where, L is the intensity of transmitted light and L_0 is the intensity of the incident light. A transparent glass was used as the blank for reference, during the light transmittance test. All the samples had uniform thickness of 1 mm. The density of the samples was determined using Archimedes method with a densitometer (New Classic MS Mettler Toledo). The hardness of the samples were determined using a Vickers hardness tester (Mikata HVS10, Japan) with 0.2 kg f load and dwelling time of 10 sec. The standard used in the hardness test was based on ASTM C 1327-03 (Standard Test Method for Vickers Indentation Hardness of Advanced Ceramics). The samples were prepared for morphology analysis according to ASTM E3-01 (Standard Guide for Preparation of Metallographic Specimens). The samples were ground with different grades of silicon carbide paper (600, 800 and 1200 grid) and subsequently polished with diamond paste (1 and 3 µm). The samples were then thermally etched at 1400°C for 1 h and characterized by field-emission scanning electron microscopy (FESEM) (Zeiss, Supra 55VP, USA).

RESULTS

Light transmittance of fabricated samples: This test measures light transmittance of the fabricated samples within the visible light spectrum, which has a wavelength range of 400-780 nm. Different wavelengths have different values of transmittance through a specimen; thus, the result shows the average value. The light transmittance results of the specimens (1 mm) ranged from 7.41-35.61% (Fig. 3). Each wt% of alumina addition seems to have a different optimum value of PEI addition that could yield the highest transmittance in the said category. However, maximum light transmittance was obtained with the sample having 0.4 wt% alumina and 0.4 wt% PEI addition (35.61%). The light transmittance of 3YSZ improved by 17.6% compared with the study by Jiang *et al.* (2011), which used 40 nm powder and uniaxial pressing to fabricate translucent 3YSZ (0.5 mm). The results still varied despite both studies employing similar theory in calculating light transmittance because the specimen thickness and the instrument setup were different. Kim *et al.* (2013), studied the effect of sintering conditions of commercial dental zirconia (Lava and Kavo) on the grain size and translucency and employed similar sample thickness (1 mm) and similar instrument setup (spectrometer). They reported that the maximum light transmittance value of

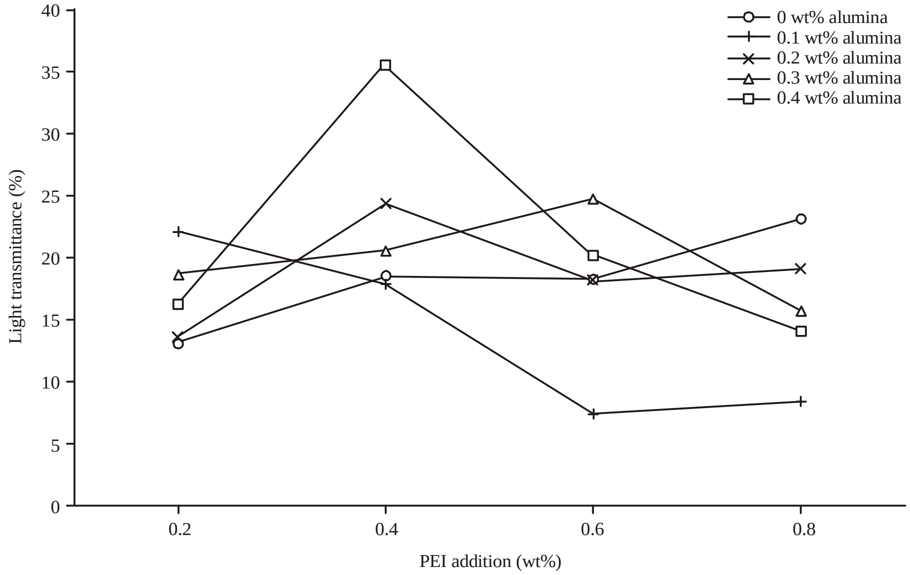

Fig. 3: Light transmittance of 3YSZ specimens with different amounts of alumina and PEI

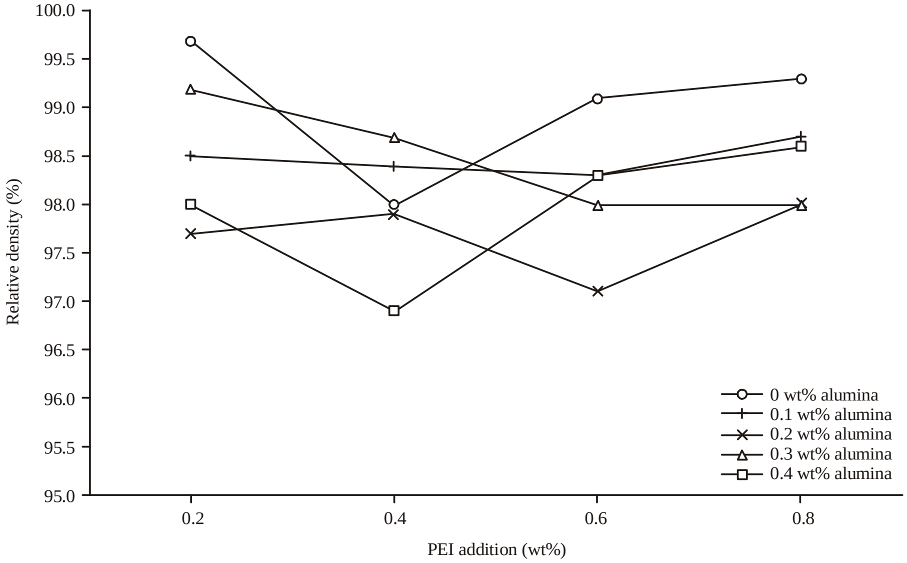

Fig. 4: Relative density of fabricated sample

Lava and Kavo were 28.39-34.48 and 28.09-30.50%, respectively. This result showed slight improvement in translucency of the fabricated zirconia samples compared with the commercially available materials.

Relative density and vickers hardness: The sintered relative density of zirconia samples is shown in Fig. 4. The relative density of the sintered samples ranged from 96.9-99.7%. The highest density (6.08 g cm^{-3}) was obtained with 0.0 wt% alumina and 0.2 wt% PEI addition into 3YSZ. Figure 5 shows the hardness of the sample with different alumina and PEI additions. The results showed that the specimens had hardness ranging from 12.7-13.6 GPa. This result is a slight

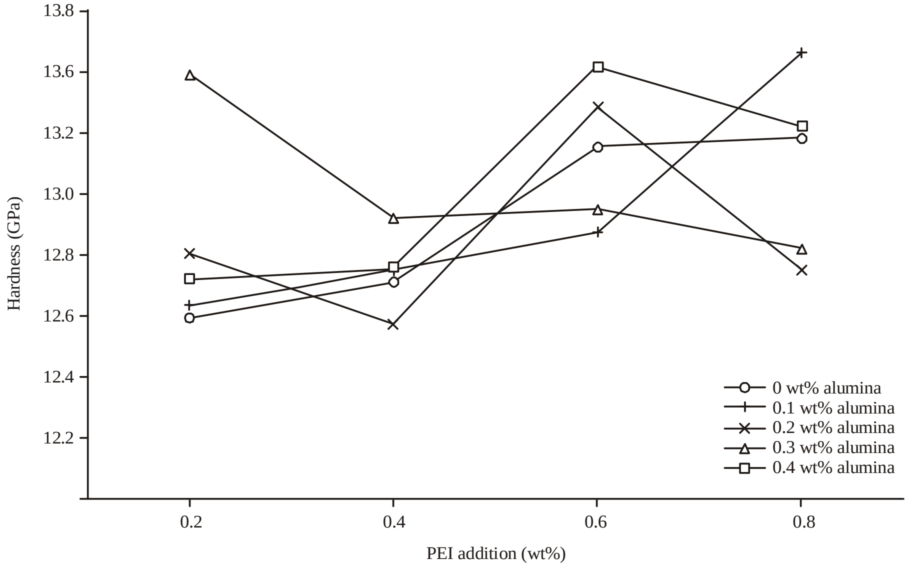

Fig. 5: Hardness of 3YSZ specimens with different amounts of alumina and PEI addition

improvement on Amat *et al.* (2014), in which the highest value of hardness is 12.9 GPa. The different combinations of alumina and PEI addition seem to have had insignificant impact on the hardness of the 3YSZ specimens, suggesting that 3YSZ can retain its mechanical strength despite having different amounts of additives in the microstructure.

DISCUSSION

The light transmittance result indicates that the translucency of 3YSZ can be improved by using alumina as sintering aid and PEI as dispersing agent. The role of PEI in fabricating translucent zirconia is important because this agent enables the creation of a green body with uniform density, which is a major concern and also because the number of light scattering centers, such as pores are kept to a minimum after sintering (Mouzon *et al.*, 2008). Although PEI aids in homogeneity, the minute addition of alumina also helped to inhibit the low-temperature degradation of zirconia by stabilizing the tetragonal phase (Chen *et al.*, 2008; Reyes-Rojas *et al.*, 2012). This phenomenon is important because the monoclinic and tetragonal phases of zirconia have different refractive indexes (Djurado *et al.*, 2000; Garcia *et al.*, 2006). The presence of two types of phases in the crystalline structure (with different refractive indexes) of zirconia can cause the light to be intensely scattered and diffusely reflected (Jiang *et al.*, 2011). Light scattering at the interface between adjacent crystals can be reduced with the formation of homogeneous tetragonal microcrystal.

The morphology of the samples was characterized by FESEM, which shows the cross section of a polished and thermally etched zirconia sample. Figure 6 shows the comparison of the FESEM micrographs of two samples: (a) A sample without any alumina addition and 0.2 wt% PEI addition, which has the highest sintered density and (b) Shows the sample with 0.4 wt% alumina and 0.4 wt% PEI addition, which has the highest translucency but low sintered density. The darker region in Fig. 6b can be attributed to the alumina rich phases, according to Energy Dispersive X-ray (EDX) data.

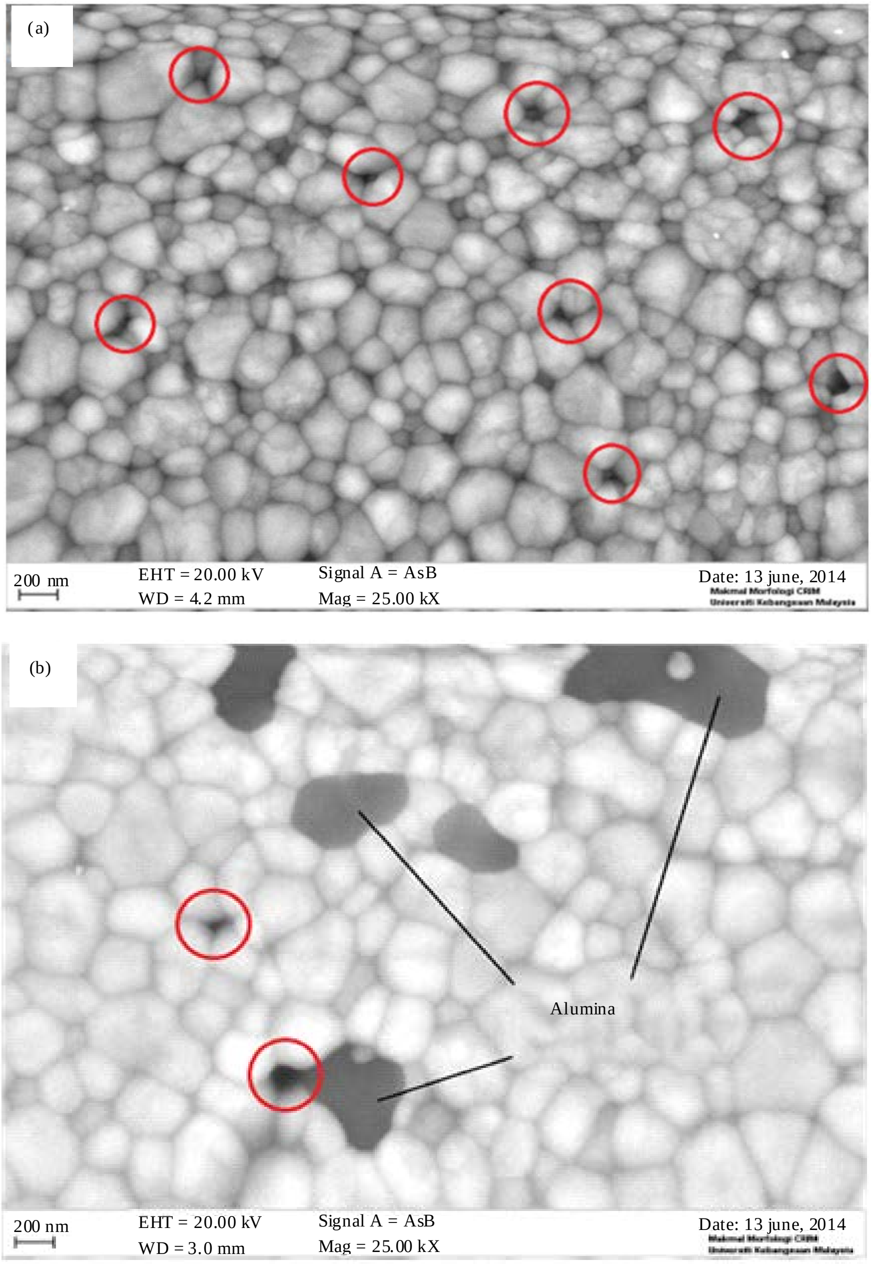

Fig. 6(a-b): (a) SEM micrograph of 0 wt% alumina and 0.2 wt% PEI sample and (b) SEM micrograph of 0.4 wt% alumina and 0.4 wt% PEI sample

The FESEM and density analyses showed that the zirconia sample without alumina addition had a smaller grain size and higher relative density. However, the grain size of sample (a) was heterogeneous, with more pores in the microstructure (as indicated in the circle). On the contrary, sample (b) with 0.4 wt% alumina and PEI had lower relative density and larger grain size but also exhibited more homogeneous grain and fewer pores. The drop in relative density is consistent with the results obtained by Chen *et al.* (2008). This result is most probably attributed to the amount of alumina rich phases, which has lower density than the tetragonal phase of zirconia. The larger grain size in specimen (b) may be attributed to the excessive grain growth during the sintering process. With the presence of alumina as sintering aid, the green bodies had reached full densification during the soaking time of sintering process. The excessive energy given during the 2 h long soaking time then causes the growth of grain size. Therefore, much research can be conducted to determine the proper soaking time to enhance the translucency of zirconia by creating a microstructure with smaller grain size.

The possible explanation behind better translucency of zirconia with larger grain size is because of the birefringence properties of zirconia. Materials with birefringence properties have anisotropic crystal structure, which exhibits different refractive indexes on different crystalline directions. The changes in refractive index will then cause scattering of the light passing through, which in turn result in lower light transmittance (Zhang, 2014). Therefore, with increasing grain size, the light that travels though the material encounters less grain boundaries hence, light scattering is minimized.

CONCLUSION

The 3YSZ specimens were fabricated with different amounts of alumina and PEI. The green bodies were slip casted and sintered at 1500°C. The specimen with the highest light transmittance of 35.61% was fabricated with 0.4 wt% alumina and 0.4 wt% PEI. However, the 3YSZ specimen with the highest light transmittance had larger grain size because of excessive grain growth and lower relative density, because of the phase, which has low density. The FESEM micrographs showed that the specimen with the highest light transmittance had fewer pores and homogeneous grain size, which lead to enhanced translucency.

ACKNOWLEDGMENT

This work was supported by the Universiti Kebangsaan Malaysia (UKM) and the Ministry of Science, Technology and Innovation, Malaysia via research sponsorship of GUP-2014-070 and 03-01-02-SF1079. The authors acknowledge the support of the Centre for Research and Instrumentation Management (CRIM), UKM for excellent testing equipments.

REFERENCES

Alaniz, J.E., F.G. Perez-Gutierrez, G. Aguilar and J.E. Garay, 2009. Optical properties of transparent nanocrystalline yttria stabilized zirconia. Optical Mater., 32: 62-68.

Amat, N.F., A. Muchtar, M.J. Ghazali and N. Yahaya, 2014. Suspension stability and sintering influence on yttria-stabilized zirconia fabricated by colloidal processing. Ceramics Int., 40: 5413-5419.

Apetz, R. and M.P. Bruggen, 2003. Transparent alumina: A light-scattering model. J. Am. Ceramic Soc., 86: 480-486.

Chang, Y.Y., 2010. Maximizing esthetic results on zirconia-based restorations. General Dentistry, 59: 440-445.

Chen, C.C., H.I. Hsiang and C.M. Zhou, 2008. Effects of the addition of minute amounts of alumina on the microstructure and sintering behavior of yttria stabilized tetragonal zirconia polycrystals ceramic via a co-precipitation process. J. Ceramic Process. Res., 9: 234-239.

Djurado, E., P. Bouvier and G. Lucazeau, 2000. Crystallite size effect on the tetragonal-monoclinic transition of undoped nanocrystalline zirconia studied by XRD and Raman spectrometry. J. Solid State Chem., 149: 399-407.

Garcia, J.C., L.M.R. Scolfaro, A.T. Lino, V.N. Freire and G.A. Farias *et al*., 2006. Structural, electronic and optical properties of $ZrO_2ZrO_2ZrO_2$ from ab initio calculations. J. Applied Phys., Vol. 100. 10.1063/1.2386967

Huo, D., Y. Zheng, X. Sun, X. Li and S. Liu, 2012. Preparation of transparent Y2O3 ceramic by slip casting and vacuum sintering. J. Rare Earth, 30: 57-62.

Jiang, L., Y. Liao, Q. Wan and W. Li, 2011. Effects of sintering temperature and particle size on the translucency of zirconium dioxide dental ceramic. J. Mater. Sci. Mater. Med., 22: 2429-2435.

Kim, M.J., J.S. Ahn, J.H. Kim, H.Y. Kim and W.C. Kim, 2013. Effects of the sintering conditions of dental zirconia ceramics on the grain size and translucency. J. Adv. Prosthodontics, 5: 161-166.

Li, J., L. Yunmao, L. Wei, W. Qianbing and Z. Yongqi, 2010. The technology of improving the optical property for the zirconia dental ceramic. Proceedings of the 3rd International Nanoelectronics Conference, January 3-8, 2010, Hong Kong, pp: 1413-1414.

Lopez-Honorato, E., M. Dessoliers, I.P. Shapiro, X. Wang and P. Xiao, 2012. Improvements to the sintering of yttria-stabilized zirconia by the addition of Ni. Ceramics Int., 38: 6777-6782.

Luo, X.P. and L. Zhang, 2010. Effect of veneering techniques on color and translucency of Y-TZP. J. Prosthodontics, 19: 465-470.

Mouzon, J., E. Glowacki and M. Oden, 2008. Comparison between slip-casting and uniaxial pressing for the fabrication of translucent yttria ceramics. J. Mater. Sci., 43: 2849-2856.

Reyes-Rojas, A., E. Torres-Moye, O. Solis-Canto, A. Aguilar-Elguezabal and M.H. Bocanegra-Bernal, 2012. X-ray diffraction and atomic force microscopy study in aged zirconia-toughened alumina composite with dispersion of m-ZrO_2 nanoparticles. Int. J. Refractory Metals Hard Mater., 35: 270-278.

Spyropoulou, P.E., E.C. Giroux, M.E. Razzoog and R.E. Duff, 2011. Translucency of shaded zirconia core material. J. Prosthetic Dentistry, 105: 304-307.

Zhang, Y., 2014. Making yttria-stabilized tetragonal zirconia translucent. Dental Mater., 30: 1195-1203.