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INFLUENCE OF SINTERING TEMPERATURE ON TRANSLUCENCY OF YTTRIA-STABILIZED ZIRCONIA FOR DENTAL CROWN APPLICATIONS

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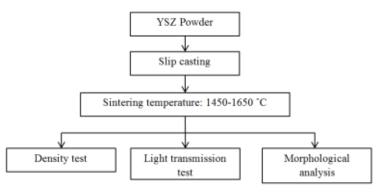
Graphical abstract



Abstract

This study aims to investigate the effect of sintering temperature on the translucency of yttria-stabilized zirconia (YSZ) for dental crown applications. YSZ suspension was treated by colloidal processing and 24 h of sedimentation to eliminate agglomerates and aggregates. The green bodies of YSZ were then shaped into pellets through slip casting. These bodies were sintered into a final shape at 1450 °C–1650 °C. The densities of the specimens were measured using Archimedes method. Light transmission of the YSZ specimen was also evaluated using a spectrophotometer with an integrating sphere. Morphological analysis was conducted with field-emission scanning electron microscopy. Results showed that sintering temperature significantly influenced the density, light transmission, and microstructure of YSZ. High sintering temperatures produced YSZ with a compact and homogeneous microstructure and a high density. Furthermore, the low light scattering effect on the porosity-free microstructure yielded light transmission as high as 37% in YSZ sintered at 1650 °C. The optimal sintering temperature was found to be 1600 °C, at which 34% light transmission was generated. In conclusion, high sintering temperatures improved the translucency of YSZ. This effect was attributed to effective densification of grains and elimination of pores at high temperatures, thereby alleviating the light scattering effect of the pores. At the optimal temperature, YSZ with high density and translucency and a compact microstructure was formed.

Keywords: Zirconia; translucency; sintering temperature; colloidal processing; slip casting

Abstrak

Kajian ini bertujuan menyiasat kesan suhu pensinteran terhadap kelutcahayaan zirkonia terstabil yttria (YSZ) untuk aplikasi korona gigi. Ampaian YSZ telah diproses melalui kaedah pemprosesan berkoloid dan pengenapan selama 24 jam untuk menyingkirkan aglomerat dan agregat. Jasad anum YSZ seterusnya dijadikan dalam bentuk pelet melalui kaedah penuangan buburan. Kemudian, jasad ini disinter pada suhu 1450-1650 °C. Ketumpatan spesimen diukur menggunakan kaedah Archimedes. Penghantaran cahaya YSZ juga disukat dengan spektrofotometer dan sfera integrasi. Analisis morfologi telah dijalankan menggunakan mikroskop elektron imbasan pancaran-medan. Keputusan menunjukkan suhu pensinteran telah mempengaruhi ketumpatan, penghantaran cahaya dan mikrostruktur YSZ secara ketara. Di samping itu, kesan penyerakan cahaya yang rendah dalam mikrostruktur tanpa liang telah membenarkan penghantaran cahaya yang tinggi iaitu 37% dalam sampel YSZ yang disinter pada 1650 °C. Suhu pensinteran yang optimum telah dikenalpasti, iaitu 1600 °C yang mana penghantaran cahaya sebanyak 34% telah diperolehi. Kesimpulannya, suhu pensinteran yang tinggi mampu meningkatkan kelutcahayaan YSZ. Kesan ini adalah disebabkan penumpatan ira dan penghapusan keliangan yang berkesan pada suhu tinggi, seterusnya meminimumkan penyerakan cahaya pada liang. Pada suhu optimum, YSZ dengan ketumpatan dan kelutcahayaan yang tinggi serta mikrostruktur yang tumpat telah dihasilkan.

Kata kunci: Zirkonia; kelutcahayaan; suhu pensinteran; pemprosesan berkoloid; penuangan buburan

1.0 INTRODUCTION

Yttria-stabilized zirconia (YSZ) has received research attention because of its superior mechanical properties. Compared with other advanced ceramics, such as alumina, YSZ exhibits extraordinary high fracture toughness (up to 10 MPa.m$^{1/2}$) and is thus named as "ceramics steel" [1]. The outstanding mechanical properties of YSZ are ascribed to transformation toughening [2], which occurs when the meta-stable tetragonal (*t*) phase of zirconia transforms into the stable monoclinic (*m*) phase. This mechanism is triggered when tetragonal zirconia is subjected to a stress field, for example, in the presence of crack propagation. *T-m* transformation is accompanied with 3%–5% volume expansion, which creates a compressive stress field around the vicinity of the crack tip. Hence, the crack tip will be closed and its propagation will be impeded. Moreover, the high biocompatibility of YSZ makes it a suitable material for biomedical applications. In the past decade, YSZ was extensively used in dental restorative applications, such as dental implantation and dental crowns. However, the insufficient translucency of YSZ mainly restricted the production of highly aesthetics dental crown [3].

Nanostructured materials have been increasingly used in several applications, such as ceramics, mainly because of their significantly improved properties compared with micro-scaled materials. Nonetheless, agglomeration occurs when using nanoscale ceramics powder as a result of increased van der Waals interparticle forces among fine particles. Agglomerates in nanoscale powder are detrimental to the mechanical properties of the final products. These agglomerates could result in heterogeneities, such as flaws, open pores, and large grains in the microstructure, which deteriorate the mechanical properties of the material [4]. Hence, agglomeration must be alleviated to generate reliable products.

Colloidal processing can effectively control agglomeration in ceramics powder. Many researchers supported that colloidal processing can manipulate interparticle forces and disperse the particles [5,6]. Colloidal processing is a wet processing method that requires ceramic particles to be dispersed in a medium, such as water, to form a suspension or slurry. The prerequisite for successful colloidal processing is high suspension stability, which can be achieved through three mechanisms, namely, electrostatic, steric, and electrosteric stabilization. These mechanisms are facilitated through pH adjustment and addition of polymers or polyelectrolytes. Slip casting is a consolidation method widely applied to fabricate ceramic products with complex geometries. This method is relatively simple and less expensive compared with other popular techniques, such as injection molding and hot isostatic pressing.

Translucency of ceramics is related to the porosity of the microstructure. Many studies proved that pores or voids in the microstructure bring significant light scattering effect, in which light passing through a pore is diffusively scattered to other directions [7,8]. Strong light scattering effect in a ceramic body yields low translucency because less light can be transmitted. In dental crown applications, translucency of the core or substructure plays an important role in matching the natural tooth of human to obtain high aesthetics value. Other factors that influence translucency of YSZ include grain size, sintering temperature, sintering method, and primary particle size.

Sintering temperature plays an important role in grain densification and thus affects the translucency of the specimen [7]. In this study, the effect of sintering temperature on the translucency of YSZ was investigated to develop highly aesthetics dental crown. Moreover, this study aims to determine the optimal sintering temperature to obtain YSZ with high translucency. High sintering temperatures are predicted to improve the translucency of YSZ.

2.0 METHODOLOGY

2.1 Preparation of YSZ Suspension via Colloidal Processing

30 g of YSZ nanopowder (3 mol% Y_2O_3; US Research Nanomaterial Inc., Houston, TX, USA) was magnetically mixed with 70 g of distilled water. The YSZ powder possesses a primary particle size of 20 nm, as shown in the transmission electron microscopy (TEM) image in Figure 1, and a BET specific surface area of 30 m^2/g. The suspension was subjected to colloidal processing to

suppress agglomeration in the YSZ nanopowder. Subsequently, 0.4 wt % 0.005 M polyethyleneimine (PEI) (Sigma–Aldrich, USA) was added to the suspension as dispersant. The pH of the suspension was adjusted to 2 by adding HNO_3 solution. The PEI amount and pH used were optimized in a previous study [9]. The YSZ suspension was immersed in an ultrasonic bath for 15 min to break soft agglomerates and then subjected to 24 h of sedimentation to segregate hard agglomerates. After sedimentation, supernatant was carefully extracted from the sediment at the bottom of the vessel.

2.2 Slip Casting and Sintering

The extracted YSZ suspension was poured into a 15 mm-diameter mold placed on top of a plaster of Paris block. The suspension was dried for 24 h and consolidated into green bodies of cylindrical disc shape. The green bodies were then sintered at different temperatures (1450 °C–1600 °C) with a soaking time of 2 h. Figure 2 shows the sintering profile of the process.

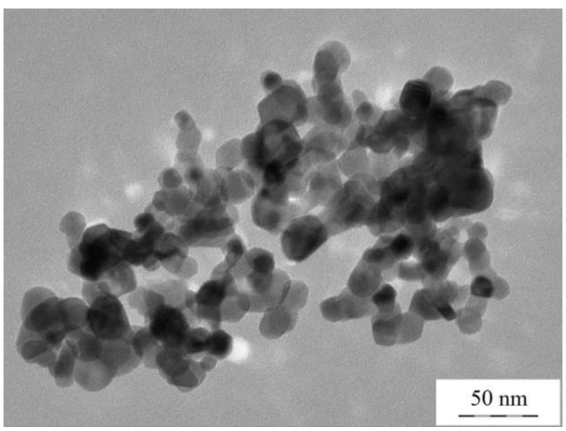


Figure 1 TEM image of YSZ powder

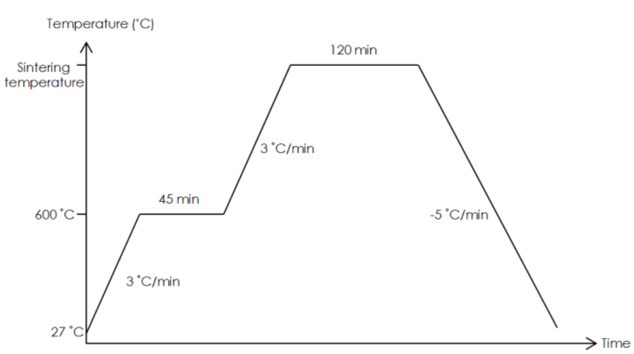


Figure 2 Sintering profile

2.3 Characterization Tests

Density of the sintered YSZ specimen was determined through Archimedes method with a density meter (Newclassic MS, Mettler Toledo, USA). Six specimens from each group were measured and the average density was obtained. The morphology of the specimen was also analyzed through field-emission scanning electron microscopy (FESEM; Hitachi SU8000, Hitachi, Japan). Prior to FESEM analysis, surface preparation was performed, in which the cross-sectional surface of the YSZ specimen was ground with silicon carbide paper (400, 600, 800, and 1200 grit) and then polished with diamond paste of 3 and 1 µ grit size. Thermal etching at 1200 °C was also required to improve grain visibility under FESEM. Finally, translucency of the specimen was characterized by determining light transmission through the YSZ body. The thickness of the specimen must be controlled at 0.5 ± 0.05 mm. A digital vernier caliper, with accuracy up to 0.01 mm, was used to measure the thickness of the specimen. Light transmission was measured with a spectrophotometer (Lambda 950, PerkinElmer, USA) with an integrating sphere (60 mm Int. Sphere, PerkinElmer, USA). Light transmission was also measured in the visible light spectrum, which included incident light with wavelength of 400–700 nm.

3.0 RESULTS AND DISCUSSION

3.1 Density

Figure 3 shows that the density of YSZ specimens increased with increasing sintering temperature. The density of the specimen reached a plateau at 1550 °C and did not significantly increase beyond this temperature. This finding indicated that the YSZ specimen achieved full densification, in which the sintered density was more than 98% of the theoretical density (6.1 g/cm^3). A maximum density of 99.5% of the theoretical density was recorded at 1600 °C.

Density is an important physical property related to the porosity of the microstructure [5]. Generally, a dense ceramic exhibits a packed microstructure with limited porosity. Pores in the microstructure are detrimental to the mechanical properties and translucency of YSZ. Hence, the density of YSZ must be measured because it indicates the porosity of the microstructure. In ceramics processing, full densification of grains is desirable to obtain high mechanical properties of the specimen.

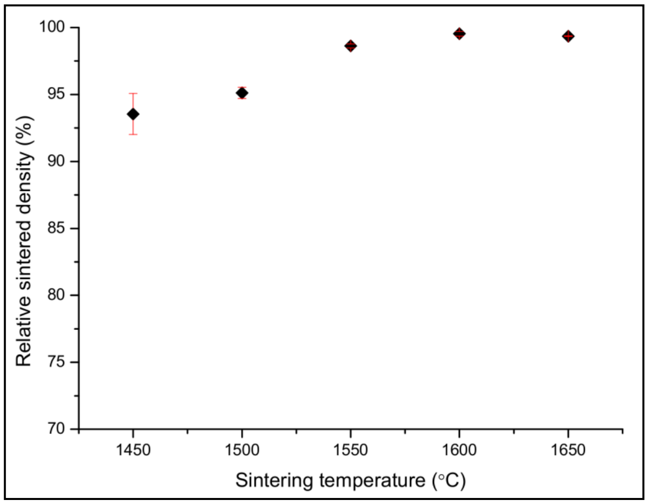


Figure 3 Density of YSZ specimens

3.2 Morphology Analysis

The microstructures of YSZ specimens sintered at different temperatures are shown in Figure 4. The porosity of the microstructure was reduced with increasing sintering temperature. At sintering temperatures of 1600 °C and 1650 °C, compact microstructures without evident pores were obtained. This finding explained the increasing density of YSZ with increasing sintering temperature. Generally, specimens with high density exhibit low porosity.

Reduced porosity with increasing sintering temperature could be attributed to high driving energy for solid-state diffusion of particles at high temperatures. Grain densification involves a massive diffusion of YSZ particles; this phenomenon requires an enormous amount of energy, specifically thermal energy. During this process, pores between the particles are eliminated as the neighbouring grains coalesce to form larger grains. If the sintering temperature is too low, then grain densification is deterred because of insufficient energy for particle diffusion. As such, many opened pores were formed in the microstructure of YSZ specimens sintered at low temperatures (1450 °C and 1500 °C).

Grain size in the YSZ microstructure increased with increasing sintering temperature. Notably, YSZ specimens sintered at 1550 °C, 1600 °C, and 1650 °C exhibit average grain sizes of 400 ± 11, 550 ± 20, and 700 ± 35 nm, respectively. Large grain sizes promote crack formation and propagation, thereby deteriorating both mechanical and optical properties of YSZ [5,10]. Muchtar and Lim [11] explained that large grains are less resistant to trans-granular cleavage, and cracks can propagate easily across large grains. Moreover, the grain size threshold for spontaneous *t-m* transformation was identified at 1 µm [12]; this phenomenon significantly reduces the mechanical properties of YSZ because of diminished transformation toughening mechanism.

In summary, the microstructure of YSZ sintered at 1600 °C exhibits optimal features to produce high translucency. The microstructure is compact and homogeneous with limited porosity. Furthermore, grain size at 1600 °C is finer than that at 1650 °C and is thus more promising in producing high mechanical properties. These features are also known to benefit the optical properties of YSZ.

3.3 Light Transmission

In this study, the translucency of YSZ was characterized with light transmission measurement. A large amount of light can be transmitted through a body with high translucency. Figure 5 shows the light transmission of YSZ specimens sintered at different temperatures. The translucency of YSZ was significantly enhanced as the sintering temperature increased from 1450 °C to 1650 °C. Moreover, YSZ specimens sintered at low temperatures exhibited an opaque optical property, with light transmission of lower than 5%. The highest translucency was found in YSZ sintered at 1650 °C with a light transmission of 37%. Sintering at 1600 °C produced high translucency with a light transmission of 34%, which is slightly lower than that at 1650 °C.

The increasing trend of YSZ light transmission with increasing sintering temperature can be explained by decreasing porosity in the microstructure with increasing temperature [13,14]. Pores are highly effective light scattering centres in ceramics [15]. The light scattering effect occurs because of significant differences in light refractive index between pores (1.00) and YSZ (approximately 2.20) [7]. As light is transmitted through a body and encounter a pore, the light will be diffusively scattered in various directions. Hence, scattering of light decreases the intensity of transmitted light and generates low translucency. When the porosity level in the microstructure is high, light will be scattered and thus yields low translucency.

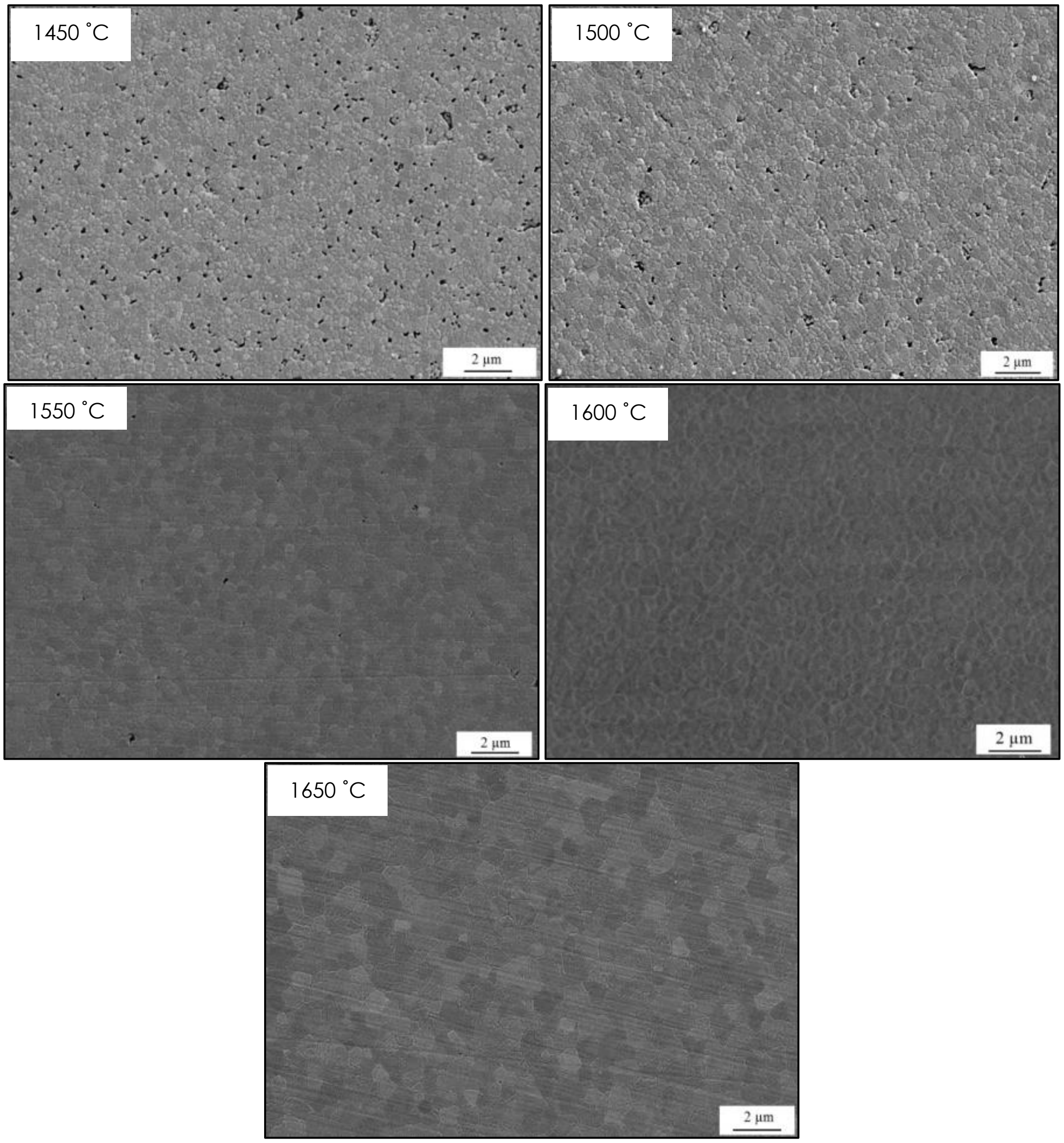


Figure 4 SEM images of YSZ specimens sintered at different temperatures

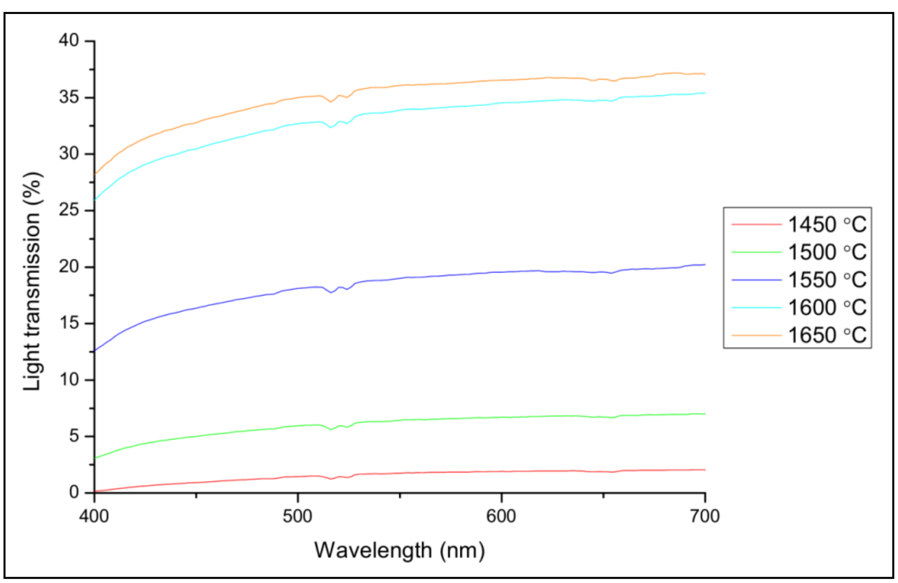


Figure 5 Light transmissions of YSZ specimens sintered at different sintering temperatures

4.0 CONCLUSION

Sintering temperature significantly influenced the translucency of YSZ. High sintering temperatures resulted in improved translucency of YSZ. This finding could be attributed to reduction of porosity in the microstructure at high sintering temperatures, eventually resulting in less light scattering effect. Hence, high translucency can be obtained as more light can be transmitted through the body. The optimal sintering temperature is 1600 °C, which produced YSZ with high density (99.5%) and translucency (34 %) and a compact microstructure.

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