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The Effect of Sintering Temperature of 8YSZ Powder to Densification and Crystal Structure

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ABSTRACT. In this paper, effect of sintering temperature on the microstructure and phase transformation in 8 mol% of yttria stabilized zirconia (8YSZ) has been studied. 8YSZ ceramic materials sintered at 1550 °C to 1700 °C were investigated. The fraction of tetragonal to monoclinic transformation increase with the increment in sintering temperature as grain size increase. Major fraction of monoclinic zirconia phase was found in 8YSZ matrix after increase sintering temperature. The fraction of cubic zirconia phase was also found increase slightly with the increase in sintering temperature. Empirical results from XRD, SEM and density clearly indicate that the highest densification and dominant phase obtained. Granulated 8YSZ presented that no significant on sintering temperature.

Keywords: Yttria stabilized zirconia, Densification, Rietveld refinement, X-ray diffraction;

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1. INTRODUCTION

Granulation is an agglomeration method that obtains granules with desired characteristics and functionality [1]. Gardner et al. [2] stated that granulated powder are good in flow ability and filling performance during compaction, thus can produce dense ceramic. Granulated powders provide rapid uniform fill to minimize part to part variations. Besides, granulated powder allows rapid and continuous release of entrapped air during compaction to avoid the formation of defects like delamination that might occur due to channelling of escaping air that to occur in non-granulated ceramics. Granulated 8 mol% of yttria stabilized zirconia (8YSZ) is one of the most common electrolytes materials used in solid oxide fuel cell (SOFC), with many advantages such as low thermal expansion coefficient (10.7×10^{-6} K^{-1}), low thermal conductivity (2.1 Wm^{-1} K^{-1}) at 1000 °C, high thermal shock resistance [3]. In order to produce dense 8YSZ ceramic, the effect of temperature needs to be systematically studied. Wang et al. [4] pointed out that sintering process induces particles to bonded together by diffusion, mass transport from the regions with the high chemical potential to lower chemical potential and neck growth between particles. After sintering, residual porosity occurred due to presence of free gaps between particles. Consequently, Ali et al. [5] had reported that sample tends to increase the bonding strength between particles as well as lower the modulus of elasticity for some ceramic. Therefore, the present research was conducted to investigate the effect of sintering temperature on the microstructure and phase stability of 8YSZ ceramic. The structure and phase stability was studied by using X-ray diffraction (XRD) while scanning electron microscope (SEM) was used to elucidate the microstructure.

2. MATERIALS AND METHODS

The work described in this paper was conducted with the same material used in a previous paper [6, 7]. Powders of 8 mol% yttria- stabilized zirconia (8YSZ) with average particle size less than 160 μm were purchased from Maju Saintifik. The particle size distribution was determined with a laser diffraction analyser (Mastersizer S, Malvern, Worcestershire, United Kingdom). Phase composition of 8YSZ powder comprised of 3.8% of monoclinic crystal structure. A sample of 3.5 g of 8YSZ and approximately 3 to 4 drops binder polyvinyl alcohol (PVA) were mixed and press into cylindrical sample (diameter = 20 mm and height = 2.4 mm) with pressure 220 MPa by using Automatic hydraulic press (3890.4NE000). Compact green body pellets were undergoes different sintering temperature 1550 °C, 1600 °C, 1650 °C and 1700 °C and hold for 4 hours, respectively. Density of the sintered ceramics were measured based on Archimedes principle, while phase composition were analyzed with X-ray diffractometer. All XRD patterns were collected at room temperature in range of 2θ = 5° to 90°. The diffraction signals were collected with step size of 0.02°. Rietveld refinement method using Xpert HighScore Plus software to quantify phase and refine the crystal structure. The crystallite size (D) was estimated from line broadening of the (111) peak. D was calculated from Scherrer equation: $D= 0.9\lambda/\beta\cos\theta$; where λ is the wavelength of the X-rays, β is the corrected full width at half-maximum of the peak and θ is the diffraction angle. Besides, the surface of the sintered specimens observed by HITACHI TM3000 Tabletop Microscope.

3. RESULTS AND DISCUSSION

Fig. 1 (a) shows that typical spectra of XRD for $(ZrO_2)_{0.92}(Y_2O_3)_{0.08}$ ceramics sample prepared at varying sintering temperatures (1550 °C, 1600 °C, 1650 °C and 1700 °C for 4 hours). The phase composition of 8YSZ ceramic only consists of tetragonal zirconia (ICSD 98-002-0789) with space group P42/nmc, cubic zirconia (ICSD 98-003-2797) with space group Fm3m and monoclinic zirconia (ICSD 98-010-8439) with space group P121/c1. The main phase composition including cubic ZrO_2 and tetragonal ZrO_2, followed by monoclinic ZrO_2. The XRD patterns reveal the absence of extra reflections corresponding to any secondary phase of impurity in the ceramics because solid solutions involving there elements have formed through inter-diffusion and the solid solubility is below than the corresponding limit. Reflections at 30°, 35° and 50° were observed in all sintered 8YSZ ceramic samples attribute to the overlapping of cubic and tetragonal phases. It was obviously display that the particular peaks involves the combination of both cubic and tetragonal phases. It is hard to single out that particular peak is refer to what phases. Thus, crystal structure refinement by using Rietveld method was used to identify the polymorphs present for each sample as display in Table 1. Moreover, it is visibly that cubic zirconia generated high intensity at sintering temperature 1550 °C. Major monoclinic phases was detected at hkl, $(11\bar{1})$ at 28° and (111) at 31.3°. The 8YSZ sintered at different temperature have same phase structures (cubic, tetragonal and monoclinic). Colomer et al. [8] stated that the highest intensity peaks corresponding to 8YSZ solid solution can be observed at (111), together with defined peaks corresponding to a cubic zirconia phase. It is well known that in cubic zirconia the coordination of Zr^{4+} ion is formed by 8 oxygen neighbors place at equal distances. The diffraction peaks with Miller indices (111) cubic phase show a shift to higher d-values (2.992) at 1600 °C whereas the peak shifted to a lower d-value (2.973) at 1550 °C. The peaks that correspond to the cubic phase are further shifted towards lower d-values.

Sintering temperature has exerted a significant influence on the typical diffraction peak positions and lattice parameters of 8YSZ [9]. From the results of qualitative phase analysis in Fig. 3(a), the intensity of diffraction pattern was not clearly revealed. When focusing at position 30° in Fig. 3(b), the intensity of diffraction pattern is the highest at 1550 °C, followed by 1650 °C, 1700 °C and 1600 °C. Correspondingly, an obvious variation in the unit cell parameters of both phases has occurred with the change in sintering temperature in ceramic. The position of the (111) diffraction peak of 8YSZ has moved to low angle region

with the increasing of sintering temperature, demonstrating the reduction of unit cell parameter of 8YSZ. It can be supported by the quantitative results as tabulated in Table 1. As presented in the Table 1, demonstrates the percentages of tetragonal (T), monoclinic (M) and cubic (C) phases of various sintering temperature (1550 °C to 1700 °C) that obtained by XRD analysis. The tetragonal phase decreasing with increment in sintering temperature. This is due to the lattice parameter started to undergo expansion when temperature arise. The cubic phase content increase from 31.3 % to 36.8 % at sintering temperature from 1550 °C to 1600 °C. This explained that the decomposition Y^{3+} ion of tetragonal 8YSZ diffused to cubic phase at high temperature [3]. A significant growth in cubic lattice parameter ascertain oxygen vacancy with increased sintering temperature. Monoclinic phase content was increasing as an increment in sintering temperature. It displays about 9.1% appears after sintered at 1550 °C and increase up to 18.5% at 1700 °C. Thus, Liu et al. [10] states high sintering temperature 1700 °C promotes phase transformation (tetragonal to monoclinic) due to volume changes in the phase stability of 8YSZ.

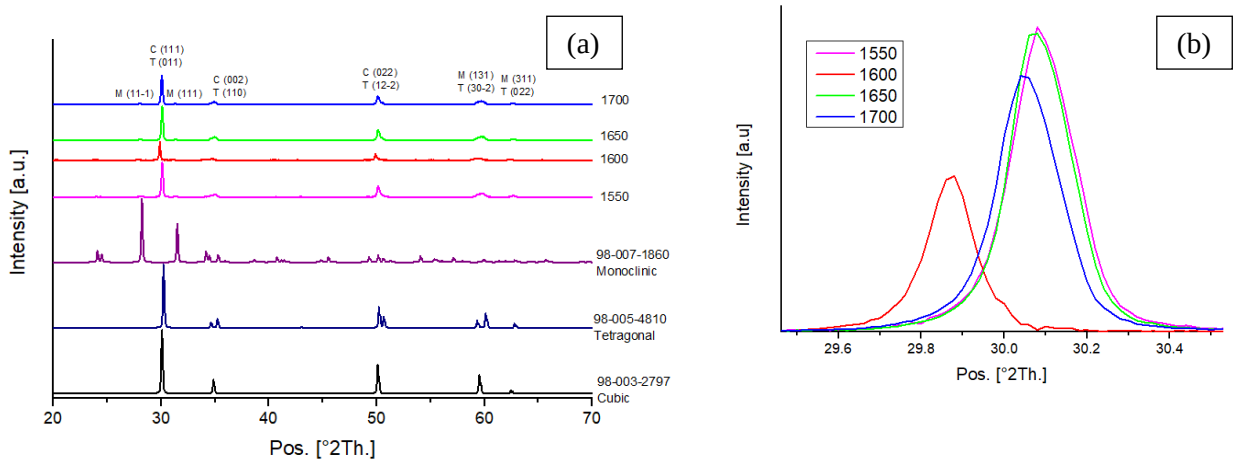

Fig. 1 (a) XRD pattern of the 8YSZ ceramics with various sintering temperature (1500 °C, 1600 °C, 1650 °C and 1700 °C) sintered for 4 hours in air and (b) comparison of dissimilar sintering temperature at position of 30°

Table 1 Percentage of phases in each sample

Sintering Temperature, °C	Tetragonal	Cubic	Monoclinic
1550	59.7	31.3	9.1
1600	50.8	36.8	12.4
1650	53.0	34.5	12.5
1700	47.3	34.2	18.5

The lattice parameters of tetragonal started to deform where c-axis undergo contraction and a-axis elongation when calcination temperatures higher than 1000 °C. Such deformation change tendency of lattice parameters as Y ion incorporate into the lattice of zirconia to form a substitution solid solution [11]. Besides that, Y ion has larger ionic radius than of Zr ion; when it incorporates into the lattice, the unit cell of zirconia expand. At temperature below 900 °C, most of the Y ion are only mixed and not a substitution of Zr atoms in the structure. It was concluded that increasing the sintering temperature suppress the grain growth. Thus, the transformation from monoclinic to tetragonal also increasing. The crystalline size for the tetragonal to

cubic phase transformation is ~6 nm from 1550 °C to 1650 °C. The crystalize size may become larger if the crystallite suffers higher lattice strain due to high sintering temperature at 1700 °C. Thus, higher the sintering temperature can lead to rapid decomposition of tetragonal 8YSZ. As a result, the effect of sintering temperature on granulated 8YSZ presented low densification (62%-67%), low percentages of cubic phase (31% - 36.8%) and eventually contributed to low ionic conductivity.

SEM morphologies of 8YSZ ceramics sintered at 1550 °C to 1700 °C for 4 hours are presented Fig. 2. The densification of the specimens was measured at different sintering temperature. The morphology shows that sample was not fully densified. Table 2 indicated that the densification of 8YSZ samples sintered at 1550 °C was about ~67.40% while the rest is only approximately ~ 62%. This variation was parallel with the SEM results depicted in Fig. 3. But then in previous research, Patil et al. [12] and Zhang et al. [13] stated that sintering temperature at 1550 °C of 8YSZ can achieve densification almost ~98%. Grain growth in Y_2O_3 stabilized zirconia will distribution of Y^{3+} ions plays a significant role in densification process. According to segregation of Y^{3+} ions on grain boundaries for 8 mol% Y_2O_3 stabilized zirconia suppresses the grain growth by a mechanism of solute drag, while the relatively uniform distribution of Y^{3+} ions for 8YSZ has a weak effect of solute drag resulting in higher grain growth rate [14]. Grain growth of 8YSZ was not achieved throughout sintering process, which Wang [4] pointed out that sintering process induces particles to bonded together by diffusion, mass transport from the regions with the high chemical potential to lower chemical potential and neck growth forms between particles. Finally, Ali et al. [5] had reported that sample tends to increase the bonding strength between particles during sintering process.

Table 2 Result of densification of 8YSZ at various sintering temperature

Sintering temperature (°C)	Bulk density (g/cm^3)	Densification (%)	Porosity (%)
1550	3.805	67.40	32.60
1600	3.585	62.26	37.74
1650	3.626	62.48	37.52
1700	3.598	62.44	37.56

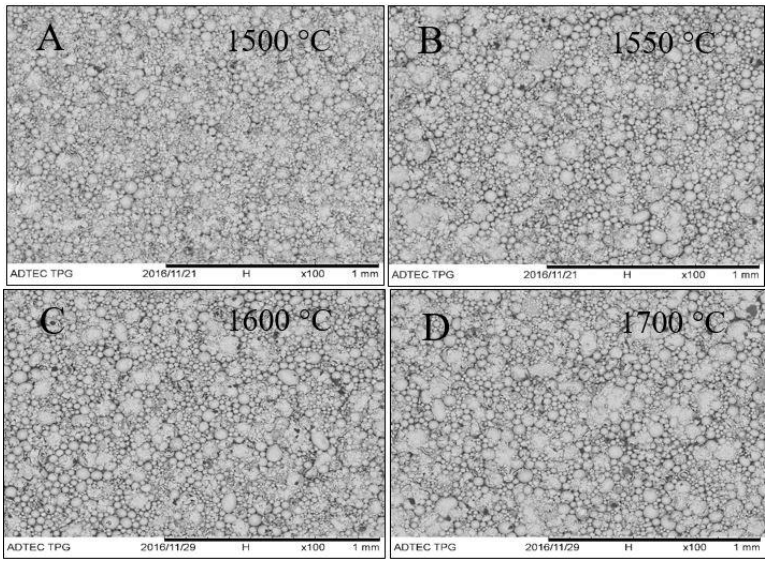

Fig. 2 Morphology of 8YSZ at different temperature (A) 1550 °C, (B) 1600 °C, (C) 1650 °C and (D) 1700 °C and hold for 4 hours

In general, surface diffusion is a matter redistribution where particles transport through the pores surface, the concentration of adsorbed particles, i.e. surface coverage is a function of complex nature of the interaction between the adsorbed particles and the surface atoms [15]. Surface diffusion is important phenomena for a sample to densify. Fig. 3 clearly shows that overall 8YSZ granulated sample was not undergo any surface diffusion between particles which cause porosity to be occurred. When surface diffusion not happen between particles, consequently lattice diffusion from the surface will not take place. It followed by grain boundary diffusion and lattice diffusion from grain boundary difficult to form between particles. Therefore, empirical results from Table 2 shown that the studied samples densified less than 95% was ascribe to huge particles size different (16-160 μm) between one another. It was demonstrated that the surface diffusion undergoes long distance transport of particles and hard to diffuse among particles.

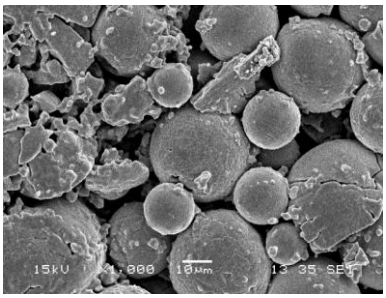

Fig. 3 Morphology of 8YSZ sintered at 1700 °C, at magnification 1000x

4. SUMMARY

Several factors such as grain size, porosity, sintering temperature and stress are responsible for densification in 8YSZ sintered samples. Although many studies on 8YSZ or ZrO_2 based material can be densified when sintered at 1550 °C, but in present study even sintered up to 1700 °C still not densified. It did not revealed that the sintering temperature is an important factor affecting the densification of granulated 8YSZ. The results showed that the granule 8YSZ strongly independent on the sintering temperature. Sintering temperature at 1550 °C to 1700 °C was lead to low densification was about 62% to 67% only. It had proven that sintering temperature does not affect significantly on 8YSZ granule particles. Further experiment will be investigated on the densification of the granule 8ysz.

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