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Spin-coated YSZ Thin Films on Silica Substrate

Shirley Tiong Palisoc, Rose Ann Tegio, Michelle Natividad, Simon Gerard Mendiola, Kevin Kaw, Stephen Tadios, Benjamin Tuason

Abstract— Different concentrations of yttria stabilized zirconia (YSZ) grown on silia (SiO_2) substrate was investiaged in this paper. Suspension containing 10wt%, 30wt%, and 50wt% YSZ were fabricated using the spin coating technique on silica keeping all other parameters constant such as the coating parameters and sintering temperature.The surface morphology and thickness of the films were investigated using scanning electron microscopy (SEM). Results showed porous YSZ films which become less porous as the concentration of YSZ increases. The thickness of the films was also affected by the YSZ concentration. As the concentration increases, the thickness of the films also increases. The crystal structure of the favricated films was also determined using X-Ray Diffraction (XRD) and Raman Spectroscopy. Both Techniques revealed a cubic fluorite structure independent of the concentration of YSZ.

Index Terms— YSZ, Raman, SEM, Spin-Coating, Thin Film, XRD

1 Introduction

WIth a 1.2% annual growth rate of world population, the total consumption of marketed energy is expected to rise by 44% from 2006 – 2030 due to the increasing demand for energy services making way for other power generation technologies to arise [1], [2]. However environmental concers should also be considered. Technology, therefore, should be able to meet the dwemands of the growing population with lwess environmental impact and more efficient use of vital energy resources. One such energy source is solid oxide fuel cell (SOFC) [3]. A solix oxide fuel cell is an electrochemical device that convers chemical energy into electrical energy [4]. Much development is focused now on SOFCs because of its high efficiency in converting a wide variety of fuels. It is also environment friendly with low emissions of NO_x and dust and since it has no moving parts and the cells are non-vibrating, noise is also eliminated [5].

Yttria stabilized zirconia (YSZ) is usually used as the electrolyte of SOFCs because of its ion conductivity, mechanical stability and good chemical compatibility. However, at operating temperatures below 800°C, the conductivity of YSZ is not high enough to lessen the resistive losses. To solve the problem, it is desirable to fabricate either thinner (<5.0μm) or more conductive films generating a power density of 400mW/cm^2 to 1500 mW/cm^2 [6].

- *Shirley Tiong Palisoc Ph.D. In Materials Science is a professor at the Department of Physics, De La Salle University – Manila, Philippines. E-mail: shirley.palisoc@dlsu.edu.ph*
- *Rose Ann Tegio is a junior officer at PNB – Manila, Philippines. E-mail: michelle.natividad@dlsu.edu.ph*
- *Michelle Natividad Ph.D. in Physics is an assistant professor at the Department of Physics, De La Salle University – Manila, Philippines. E-mail: natividad@dlsu.edu.ph*
- *Simon Gerard Mendiola is a test engineer at Hitachi Philippines. E-mail: sg_mendiola@yahoo.com*
- *Kevin Kaw is a senior student at De La Salle University – Manila, Philippines. E-mail: kevinkaw08@yahoo.com*
- *Stephen Tadios is a senior student at De La Salle University – Manila, Philippines. E-mail: stephentadios@yahoo.com*
- *Benjamin Tuason is a senior student at De La Salle University – Manila, Philippines. E-Mail: benjamintuason@yahoo.com*

Previous Studies have reported several techniques in fabricating YSZ thin films [7], [8], [9], [10]. In this study, spin coating technique is utilized because of the simplicity of its operational costs as compared to the other techniques mentioned. This study also seeks to determine the optimal condition in fabricating the films by varying the YSZ concentration and the type of substrate.

Different concentrations of YSZ namely 10wt%, 30wt%, and 50wt% are used to produce thin films on silica (SiO_2). The YSZ concentration is varied to provide baseline information on how the sample behaves within such concentration range. The sample films are characterized in terms of their surface morphology and thickness through Scanning Electron Microscopy, crystal structure though X-Ray Diffraction, and Raman Spectra though Raman Spectroscopy.

2 Experimental

2.1 Fabrication of YSZ thin films

Commercial yttria stabilized zirconia (YSZ8-U1, fuelcellmaterials.com) and ethanol (95% ethyl alcohol, Aced Laboratory) with wiegth rations of $10_{YSZ}:90_{ethanol}$, $30_{YSZ}:70_{ethanol}$ and $50_{YSZ}:50_{ethanol}$ were mixed to form a suspension. To ensure the consistency of the suspension, a Sonucator Ultrasonic Processor (Misonix) was used. The suspension was mixed for three hours with 60% output amplitude/instensity. It was then deposited on the substrate using the Spincoat G3P-8 spin coater. The substrate with YSZ layer was baked in a furnace (Thermolyne type 48000) at a temperature of 300°C for three minutes until all of the solvent evaporates. In this paper, the substrate was only coated once with the YSZ suspension. The substrate with the YSZ layer was then sintered at 650°C for four hours in the furnace.

2.2 Characterization

Using Scanning Electron Microscopy (SEM)(Jeol 5310), the surface morphology and the thickness of the films were investigated. The crystal structure of the fabricated films was determined using X'Pert PRO Pananalytical X-Ray Diffractometer (XRD) with a CuK-α radiation. Raman Spectroscopy was also utilized in this study. R3000 Raman spectrometer with 785nm laser was used to acquire the Raman spectra of the

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films. Based on the Raman spectrum, the crystal structure of the sample was obtained and was compared to the XRD result.

3 Results and Discussion

3.1 Surface morphology

Shown in Fig. 1 are the SEM images of the surface of the films fabricated on Silica substrate using three different concentrations of YSZ namely 10wt%, 30wt%, and 50wt%. Porosity of the films was evident based on the results. The film with 10wt% YSZ was highly porous that only a fre parts of the substrate were covered with YSZ (lighter portions in the image correspond to YSZ while the darker portions correspond to the substrate). As the concentration of YSZ increases *30wt% and then 50wt%), more parts of the substrate were being coated which means that the pores were getting smaller. The porosity of the films is due to the fact that YSZ poweders do not dissolve in ethanol. The function of ethanol as the suspension medium is to suspend and disperse the particles, not to dissolve them.

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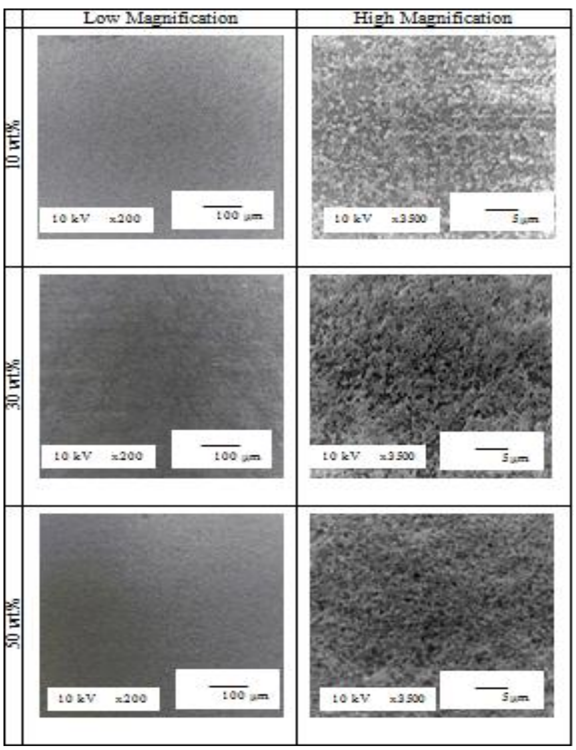

Fig. 1 SEM images of the surface of the films with different concentrations of YSZ on Silica Substrate

3.2 Film Thickness

Fig. 2 shows the SEM images of the films fabricated on Silica showing the film thickness. Thickness of 0.764µm (average thickness), 0.983µm, and 2.47µm were achieved for films with 10wt%, 30wt%, and 50wt% YSZ respectively. It is seen that, as the concentration of YSZ increases, the thickness of the film also increases which indicates that film thickness was directly proportional to the concentration of YSZ. The effecto f cutting was clearly shown especially in Fig. 2b wherein some parts were raised and others were adhered to the substrate.

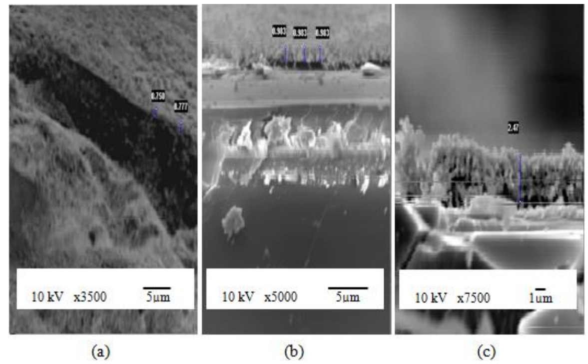

Fig. 2 SEM images showing the thickness of the films with (a) 10wt% YSZ, (b) 30wt% YSZ, (c) 50wt% YSZ fabricated on Silica.

Listed in Table 1 are the film thicknesses of the different concentrations. Since the suspension with 10wt% YSZ was runny or less viscous, it was easily spun out of the substrate thus, making the film thinner. On the other hand, the suspension with 50wt% YSZ id very viscous thus was not easily spun out of the substrate during spin coating process making the film thicker.

Table 1
Thickness of Films (in µm)

10wt% YSZ	30wt% YSZ	50wt% YSZ
0.764	0.983	2.47

3.3 Crystal Structure

3.3.1 X-Ray Diffraction results

The crystal structure of fabricated YSZ thin films was determined by x-ray diffraction. Fig. 3 shows the x-ray diffraction patter for 10wt% YSZ film fabricated on silica substrate. In X-Ray Diffraction technique, the position of the peaks indicates the crystal structure of the material while the intensity of the peaks depends on the material distribution in the structure [11]. Therefore, in determining the crystal structure of YSZ films, the focus is on the peak position. The peaks observed (30.17°, 34.97°, 50.17°, 59.71°, and 62.69°) are comparable to the XRD results obtained in the study conducted by Priyatham and Bauri [12] which signifies that the 10wt% YSZ film fabricated on Silica substrate aslo has a cubic fluorite structure. The XRD results of films with 30wt% and 50wt% YSZ were not anymore presented since the material used (YSZ) was the same for all concentrations and thus, will generate the same XRD pattern.

X-Ray Diffraction analysis revealed that the sample films obtained in this study, exhibit a cubic fluorite structure.

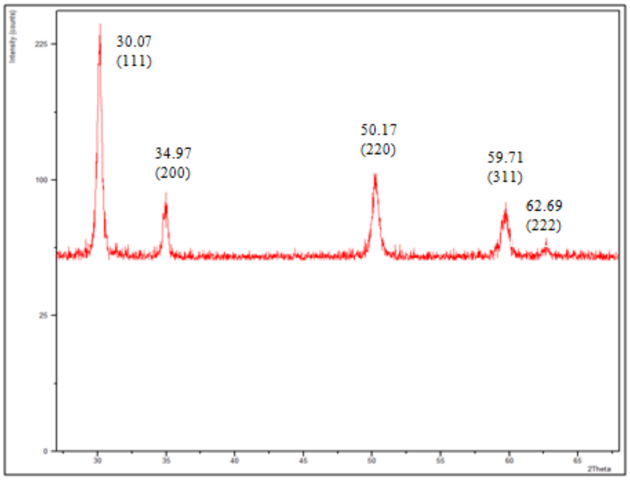

Fig.3 XRD patterns of films with 10wt% YSZ fabricated on Silica Substrate

3.3.2 Raman spectroscopy results

To verify XRD results, another test was done which was the Raman spectroscopy. The attained Raman spectra were compared to the results obtained by Ghosh et.al. [15] and Cheng and Liu [16]. Shown in Fig. 4 are the Raman Spectra of the films containing 10wt%, 30wt%, and 50wt% YSZ fabricated on silica substrates respectively with the characteristic peaks (enricled peaks) for cubic fluorite YSZ.

From the Raman spectra of films, it is observed in Fig. 5 and Table 2 that the intensity of the peaks increases with ncreaseing concentration of YSZ, in agreement with Umback and Hines [17].

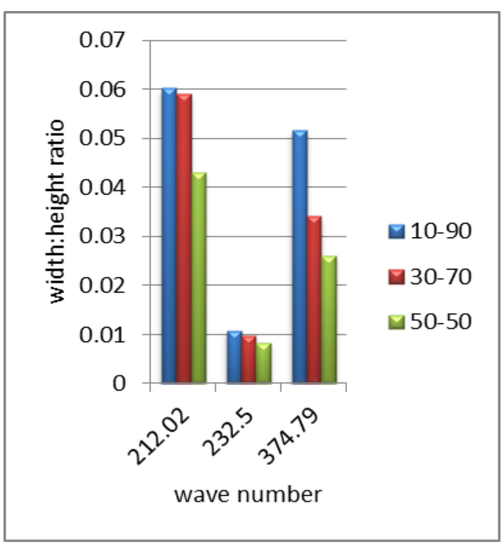

Fig. 5 Width to height ratio of Raman peaks of 10wt%, 30wt%, and 50wt% YSZ films fabricated on Silica substrate

Table 2
With to height ratios of Raman peaks of 10wt%, 30wt%, and 50wt% YSZ films on silica substrate

10 wt% YSZ		30 wt% YSZ		50 wt% YSZ	
Wave no.	Width to height	Wave no.	Width to height	Wave no.	Width to height
212.02	0.6027	212.02	0.05894	212.02	0.04297
232.5	0.01071	232.5	0.009706	232.5	0.008306
374.79	0.05164	376.16	0.03410	373.42	0.02589

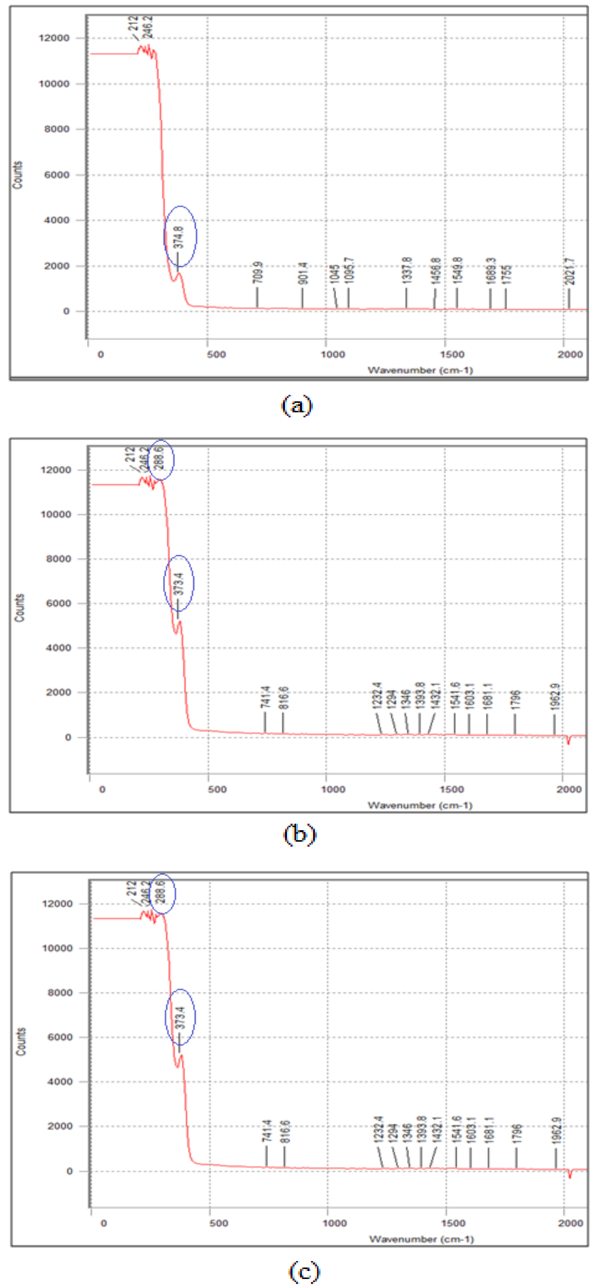

Fig. 4 Raman spectras of films with (a) 10wt% (b) 30wt% (c) 50wt% YSZ fabricated on Silica substrate

4 Conclusion

SEM images of the surface of the fabricated films showed that the films are porous. The porosity depends on the concentration of YSZ used in fabricating the films. Greater concentration means more amount of YSZ that can cover up the pores and thereby lessening the porosity of the films.

Viscosity of the suspension used was the main factor in determining the thickness of the films. Films with 10wt% YSZ are relatively thinner compared to the films with 30wt% YSZ and much thinner as compared to films with 50wt% YSZ.

X-Ray Diffraction patterns show that crystal structure of the fabricated thin film is cubic fluorute which coincides with the result obtained by Priyatham and Bauri [12]. A cubic fluorite structure indicates that the films are stable. The intensity of the peaks gets higher as the concentration of YSZ gets larger.

Similar results were observed from the Raman patters of the fabricated films. The intensity of the peaks get higher as the concentration of YSZ increases. Raman spectra of the films obtained in this study contain the characteristic peaks of cubic fluorite structure YSZ which agree with the results obtained by Ghosh et.al. and Cheng and Liu [16]. Raman spectra of the films above 1000cm^{-1} were also observed in this study.

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