JRI

Journal of Research & Innovation

Fabrication and Characterization of TiO_2 Thin Films Deposited on Glass Substrate by Spray Pyrolysis Technique

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***Abstract* – In this study, titanium dioxide (TiO_2) thin films have been deposited onto glass substrates by spray pyrolysis technique. The as-prepared TiO_2 thin films were sintered at different temperatures (T= 350°C, 400°C, 450°C, 500°C, 550°C).The phase formation and structural properties of TiO_2 thin films were studied by X-ray diffraction. XRD profiles showed that the films were monocrystalline and anatase phase. The surface morphology of TiO_2 thin films were characterized by using Scanning Electron Microscopy (SEM). According to experimental results, TiO_2 thin films can be used as the electron transporting layers for perovskite solar cell.**

Keywords: TiO_2 thin films, Spray pyrolysis method, XRD, SEM

I. INTRODUCTION

Thin film of titanium dioxide (TiO_2) has generated a lot of interest because of its attractive properties such as wide band gap, high refractive index, high dielectric constant, absence of nontoxicity and also it is considerable interest for both scientific and technical applications. Titanium dioxide or titania (TiO_2) was first produced commercially in 1923. It is obtained from a variety of ores. The bulk material of TiO_2 is widely nominated for three main phases of Anatase, Rutile and Brookite. Among them, the TiO_2 exists mostly as Rutile and Anatase phases which both of them have a tetragonal structure. However, Rutile is a high-temperature stable phase and Anatase is formed at a low temperature. The anatase phase has formed in the temperature range between 400°C -700°C and after 700°C the rutile was formed. TiO_2 thin films have been widely investigated for various optical applications and have gained considerable attention for optical fiber, gas sensor, ceramic membranes, photocatalysts and solar cell electrodes [1- 6]. A film formation of TiO_2 can be carried out by various methods such as sol-gel, spray pyrolysis, sputtering, pulsed laser deposition and electrodeposition. Among these techniques, spray pyrolysis is a sample, cost-effective technique for large area deposition and also excellent method for the deposition of metallic oxides thin films. Structural properties of TiO_2 depend on a large extend on the deposition technique. The present study is focused on structural properties of titanium dioxide thin films prepared by spray pyrolysis technique.

II. MATERIAL AND METHOD

Sample Preparation of TiO_2

For the sample preparation, corning glass slides had been cleaned with hydrochloric acid (HCl) and Distilled water in the ratios 1:10 for 30 minutes. The flow diagram of the sample preparation procedure of TiO_2 thin films by spray coating is as shown in Figure 1.To get more clean, the corning glass slides rinsed in distilled water for about 2 minutes. At room temperature (30°C), TiO_2 (2 g) and ethanol (20 ml) were mixed in the 200 ml beaker. For deposition, 2g of TiO_2 was dissolved with 20 ml of ethanol in the 200 ml beaker and then the mixture was maintained at a temperature of 70°C with magnetic stirrer for 1hr. This mixture formed precursor which was sprayed through spray gun using air as carrier gas with a pressure of 200kPa. The spray nozzle to substrate distance was set to about 20 cm with direction angle ~45°. The substrate temperatures was set to 350°C, 400°C, 450°C, 500°C, 550°C for different sample at 30 min. After the deposition, the films were allowed to cool slowly at room temperature. The structural characterization of the deposited films were carried out by X-ray diffraction (XRD) using RIGAKU, SMART Lab (monochromatic Cu-K_α radiation (λ =1.54056 Å). The XRD patterns were recorded in 2θ intervals from 10° to 70°.The surface morphology of TiO_2 thin films were characterized by using JEOL JSM-5610 LV Scanning Electron Microscopy (SEM).

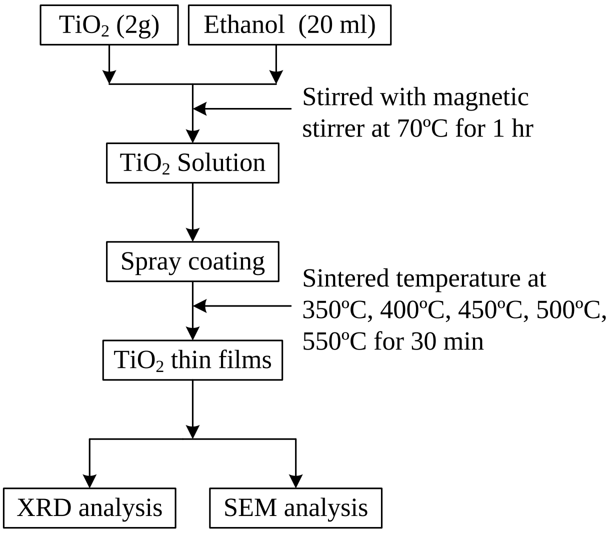

Figure 1. Block diagram of the sample preparation procedure of TiO_2 thin films by spray coating

III. RESULTS AND DISCUSSION

A. *X-ray Diffraction (XRD) Analysis of TiO_2 thin films*

XRD analysis was carried out to study the crystal structure and properties of TiO_2 thin film samples in different temperatures. It was performed using monochromatic Cu-K_α radiation (λ =1.54056 Å) operated at 40 kV (tube voltage) and 50 mA (tube current). Specimen was scanned from 10º to 70º in diffraction angle, 2θ with step-size of 10º/min. The reference profile was used as 99-0008> TiO_2-Anatase, JCPDS library files to identify the observed XRD spectral lines.

Figure 2 represented peak comparison of XRD patterns for TiO_2 thin film fabricated by using spray pyrolysis method at varying temperature. The lattice parameters of TiO_2 are a = b = 3.80 Å, c = 9.49 Å with the tetragonal symmetry. The lattice distortion (c/a) of TiO_2 is 2.496. Crystalline size of TiO_2 thin films for all identified peaks which were calculated by using the Debye- Scherrer's equation;

$$D = \frac{k\lambda}{B\cos\theta}$$

where, D is the crystallite size, k is the Scherrer's constant (0.899), λ is the wavelength of X-ray (1.54056 Å), B is the full width at half maximum of the diffraction peak, and θ is the Bragg diffraction angle.

The experimental results of the calculated average lattice parameters and the observed lattice parameters and c/a are tabulated in Table 1. Average lattice constant and crystallite size (nm) of TiO_2 thin films at different temperature were observed and tabulated in Table 2.

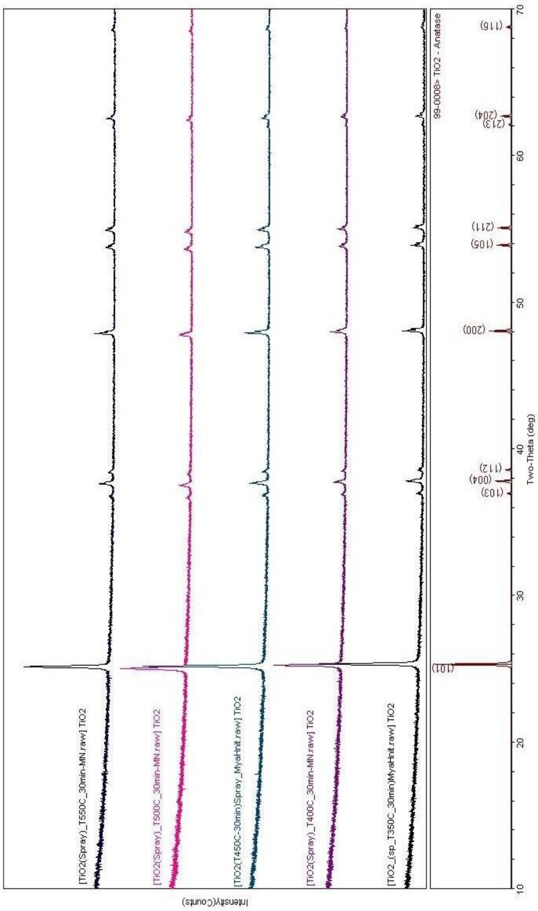

Figure 2. Peak comparison of XRD patterns for TiO_2 thin films with varying Temperature

Table 1. The lattice parametrs, and c/a of TiO_2 thin films at Different Temperatures

Temperature (ºC)	Observed Avg: a=b (Å)	Calculated Avg: a=b (Å)	Observed Avg: c (Å)	Calculated Avg: c (Å)	Calculate: c/a
350	3.79	4.05	9.45	9.51	2.35
400	3.80	3.79	9.50	9.50	2.81
450	3.81	3.80	9.49	9.55	2.51
500	3.83	3.83	9.48	9.58	2.50
550	3.81	3.80	9.52	9.55	2.51

Table 2. Average lattice constant and crystallite size(nm) of TiO_2 thin films at Different Temperature

Temperature (°C)	Avg: a=b (Å)	Avg: c (Å)	Crystallite size(nm)
350	3.79	9.45	47.50
400	3.80	9.50	45.77
450	3.81	9.49	47.23
500	3.83	9.48	49.10
550	3.81	9.52	46.89

B. *SEM Analysis of TiO_2 thin films at Different Temperatures*

Microstructural properties of TiO_2 thin films at different temperatures were characterized by using JEOL JSM- 5610 LV Scanning Electron Microscope. Figure 3 (a-e) shows the SEM analysis of TiO_2 thin films at different temperatures exhibit grained microstructure with small crystallite size. The grain sizes were calculated by using well known bar code system. Bar code size was 2 μm with magnification of 10 k . The average grain size of TiO_2 thin films were found to be 0.51 μm, 0.42 μm ,0.36 μm, 0.32 μm and 0.30 μm respectively at 350 °C, 400 °C, 450 °C, 500 °C and 550 °C . These figures indicated that most of the grain size was regular structure and a few number of large grain size were found. It looks fairly dense and rough. These consisted of well-defined grain but marked difference. This fact indicated that structural properties were influenced by different temperatures. From the images, it was clearly found that the little amount of pores and grain growth were examined with the increase in process temperatures. The orientation of grain was towards left for all images. The grain size of TiO_2 thin film at 350 °C and 400 °C were seen to be uniform but some of the grain size were slightly large and agglomerate structure from detail analysis of Figures 3 (a-e) showed the dense particle with fine grains, smooth and without any cracks sintered at 450 °C , 500 °C and 550 °C. The average grain size of the film was the largest at process temperature at 350 °C and the smallest at process temperature at 550 °C. In these results, the average grain size of the TiO_2 thin films were depended varying process temperatures.

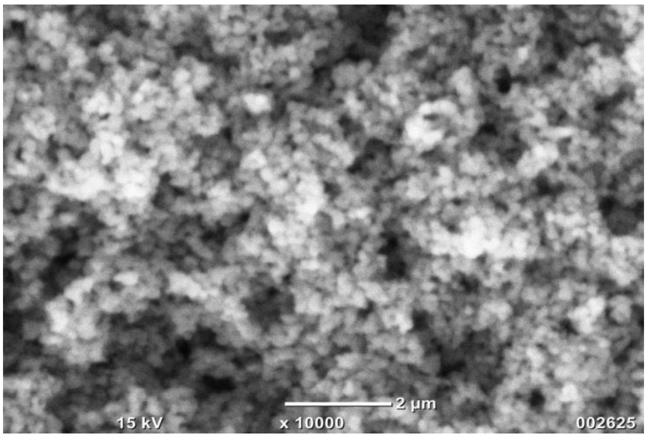

Figure 3 (a) SEM image of TiO_2 thin film at 350 ºC

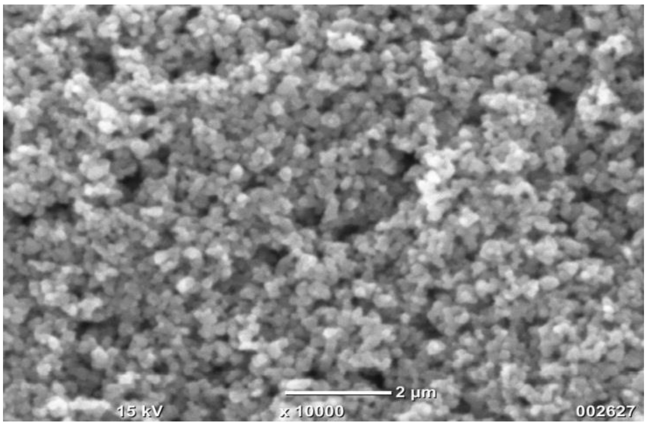

Figure 3 (b) SEM image of TiO_2 thin film at 400 ºC

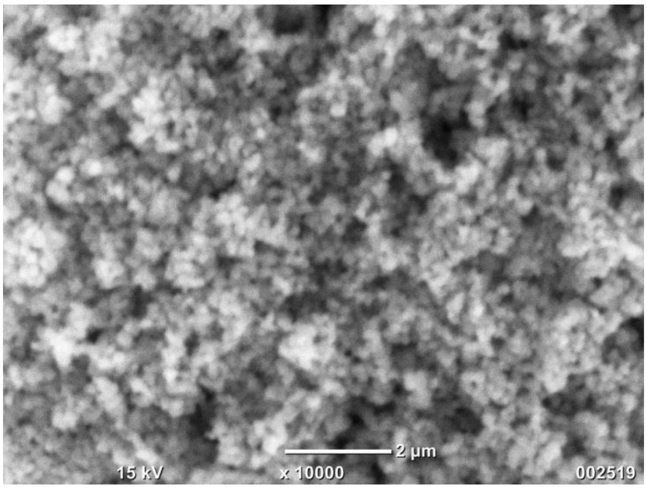

Figure 3 (c) SEM image of TiO_2 thin film at 450 ºC

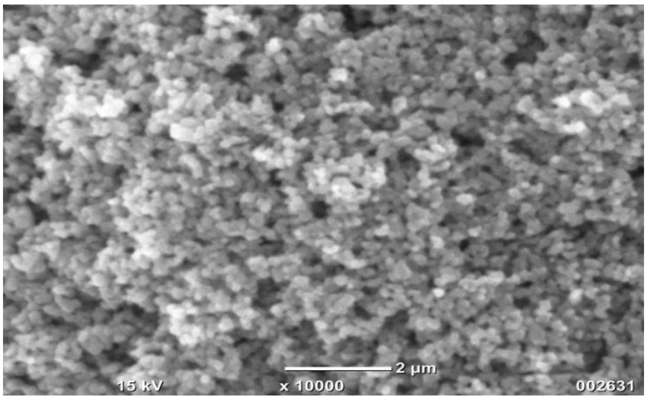

Figure 3 (d) SEM image of TiO_2 thin film at 500 ºC

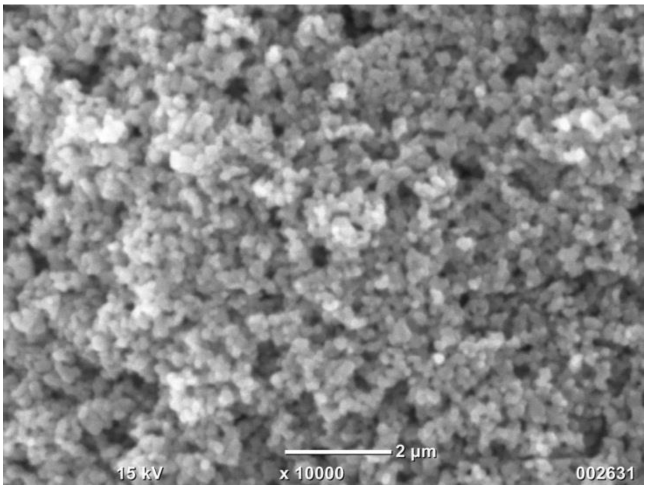

Figure 3 (e) SEM image of TiO_2 thin film at 550 ºC

IV. CONCLUSION

In this research work, the fabrication and characterization of TiO_2 thin films have been deposited on glass substrate by spray pyrolysis method at different substrate temperatures. According to XRD analysis, all the peak heights and peak positions of different temperatures were in good agreement with library files of XRD machine. The XRD pattern revealed that these films were anatase tetragonal structure. The lattice parameters of a and b were obtained in the range 3.79 Å to 3.83 Å and c was obtained in the range 9.45 Å to 9.52 Å. The crystallite size of the samples also varies between 17.10 nm to 75.90 nm. The smallest crystallite size of TiO_2 film was 45.77 nm at 400°C. From SEM images, SEM analysis was examined to be smooth, uniform, crack free and agglomerated spherical particles of TiO_2 thin films. This suggested that agglomeration of grain decreases with increasing process temperatures. The experimental finding resulted from this research work indicated that the crystal structure, phase formation and surface morphology of TiO_2 thin films were influenced by annealing different temperatures. Therefore, this TiO_2 films sintered at 400°C can be used as the electron transporting layers for perovskite solar cell.

ACKNOWLEDGEMENT

I would like to thanks Professor Dr Khin Khin Win, Head of Department of Physics, University of Yangon, for her kind permission to carry out this work. I deeply thank for my supervisor Dr Zin Min Myat, Assistant Lecturer, Department of Physics, University of Yangon, for her valuable guidance and supervision in this works.

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