Preliminary Study in Fabricating Fluorine-doped Tin Oxide by Using Spray Pyrolysis Methods

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Abstract. In this study, Fluorine-doped Tin Oxide (FTO) was fabricated by using spray pyrolysis methods for solar cell applications. The synthesis of FTO has been conducted under spray pyrolysis methods in which a precursor solution of SnCl₂: F having a concentration of 0.1, 0.3, and 0.9 M. The spray pyrolysis was performed using a nebulizer with a deposition time of 2, 3, 5 and 10 minutes. The glass substrate was heated at a temperature of 400°C prior to deposition. By controlling concentration of SnCl₂: F, FTO glass was successfully produced. The lowest sheet resistance of the FTO glass was about 60 Ω/cm². X-Ray Diffraction and Scanning Electron Microscopy were employed to investigate the characteristic of FTO. Then, FTO was applied as an electrode in the dye sensitized solar cell (DSSC) electrode application and the photovoltaic effect was observed.

Keywords: FTO, spray method, dssc application.

1. Introduction

Until now, the application of a Transparent Conductive Oxide (TCO) is very wide [1-4]. An electronic device, such as solar cells [1-3], water splitting [4] and light emitting diodes [5], were developed by applying TCO. However, the price of these components is still very expensive. It is because the method used to create TCO requires complicated production costs [6-7].

One type of TCO is Flourine doped Tin Oxide (FTO). According to Ruso (2008), that FTO is an ideal candidate for applications requiring TCO due to its ability to adhere strongly to glass, resistance to physical abrasion, chemical stability, high optical visible transparency, and electrical conductivity [8]. FTO can be produced through spray pyrolysis methods [7-9], sol gel methods [10, 11], chemical vapor deposition [12] and DC magnetron sputtering [13]. Most of the researcher using spray pyrolysis method because it is required low cost material and may produce at low temperature [8]. In this preliminary study, we made a Flourine doped Tin Oxide (FTO) by employing spray pyrolysis methods then it was applied as Dye Sensitized Solar Cells (DSSC) component. Furthermore, the performance of DSSC was performed under sun illumination.
2. Material and Methods
In this study, the materials used for the fabrication of the FTO glass were the solution of Tin(II) Chloride (SnCl\(_2\).2H\(_2\)O, Merck, Germany), Ammonium Fluoride (NH\(_4\)F, Merck, Germany), and Ethanol (96%, Merck, Germany). The SnCl\(_2\) and NH\(_4\)F materials of 1 gram each were fed into the ethanol solvent then stirred until homogeneous. Then, the spray pyrolysis method is used to deposit the solution on a glass substrate (1 cm x 1 cm). The concentration of solution and time deposition of nanoparticle are investigated to get the optimum resistance. After that, the sample was heated at 400°C for 30 minutes. A conductive transparent thin layer is then formed and conductivity measurements are made. Scanning Electron Microscopy was done to investigate the morphology of FTO film. Then, XRD diffraction is also employed to characterize the crystalization of FTO film.

3. Results and Discussion
Fabrication of FTO was performed by spray pyrolysis method (Fig. 1) [14]. It this method, nanoparticles were produced by spraying droplets from the solution onto the heated surface (Fig 1a). When a droplet contacts to a hot surface (in Fig. 1b), it will vaporize and the lagging oxide particles are then attached to the surface [9]. There are several parameters in affecting FTO resistance. It is solution concentration, time deposition, temperature and distance between nebulizer to the substrate [7].

![Figure 1. FTO fabrication process by using spray pyrolysis method: (a) Diagram of FTO deposition; (b) Process of the nanoparticles formation.](image)

In this study, to obtain a good FTO resistance, optimization of two parameters was done by varying concentration of solution and deposition time. Effect of both parameters to sheet resistance was shown in Table 1.

| No | Concentration (M) | Time Deposition (minute) | Sheet Resistance (\(\Omega\)) |
|----|------------------|--------------------------|-----------------------------|
| 1  | 0.9              | 2.0                      | 9.66 x 10^6                 |
| 2  | 0.9              | 5.0                      | 1.07 x 10^6                 |
| 3  | 0.9              | 10.0                     | 1.62 x 10^6                 |
| 4  | 0.3              | 5.0                      | 65.6                        |
| 5  | 0.3              | 10.0                     | 99.3                        |
| 6  | 0.3              | 10.0                     | 324.6                       |
| 7  | 0.1              | 3.0                      | 1.27 x 10^3                 |
| 8  | 0.1              | 10.0                     | 3.25 x 10^3                 |

From the results obtained, as shown in Table 1, it is known that both parameters (solution concentration and deposition time) affect the resulting FTO resistance. The resistance value of 65.6 to 324.6 \(\Omega/cm^2\) was found by combining the parameters. The good resistance was obtained at a concentration of 0.3 M and a 5-minute of deposition time. It was predicted that it was related to the loading level of nanoparticle formation on the substrate [15].
In order to know the crystallization and morphology of the FTO film, then the FTO sample was characterized by employing X-Ray Diffraction Spectroscopy (XRD) and Scanning Electron Microscopy (SEM).

![Figure 2](image1.png)

**Figure 2.** Characterization of FTO film: (a) XRD pattern of FTO films after annealing at temperature 400 °C; and (b) Scanning Electron Microscopy result.

Figure 2(a) shows an XRD pattern on the films after annealing at 400 °C for 30 min. The crystalline structure of the FTO films was tetragonal SnO$_2$. On the other hand, FTO nanoparticle grain can be observed at scanning electron microscopy result of FTO film in Figure 2(b). Furthermore, resulting FTO was applied as solar cells component. FTO was used as main and counter electrode of DSSC as shown in Figure 3(a). The performance of solar cells under the sun illumination can be seen in Figure 3(b).

![Figure 3](image2.png)

**Figure 3.** (a) DSSC solar cell with created FTO; (b) The measurement of DSSC under sun illumination.

The phenomenon of photovoltaics in DSSC was observed on 37.48 mW/cm$^2$ and IV characteristics were plotted in Fig. 4. Although the resulting performance of DSSC was still lack (in microwatt order) however FTO resulted have been able to be used as DSSC components as reported reference [16-18]. Furthermore, the researcher will make an efforts to obtain a better FTO in terms of conductivity, homogeneity, and transparency as suggested by the reference [16-18].
4. Conclusion

In this study, the conductive thin layer of FTO with a good conductivity about 66 \( \Omega/cm^2 \) has been successfully made using spray pyrolysis method. In the fabrication process, it was optimized concentration and time deposition. Crystallization and morphology of FTO were confirmed by employing XRD and SEM. The resulting FTO has also been tested as a DSSC component and the phenomenon of photovoltaics can be observed.

5. References

[1] Chiba, Y., et. al. 2006 Japan. J. Appl. Phys. 45(25), pp. L638–L640.
[2] Dai, S., et. al. 2005 J. Sol. Energy Mat. Sol. Cells 85, pp. 447–455.
[3] Gratzel, M., 2001 J. Sol-Gel Sci. Tech. 22 pp. 7–13.
[4] Noh E., et. al. 2013 Sci. World J. 2013 pp. 1-8.
[5] Chuang S-H, Tsung C-S, Chen C-H, Ou S-L, Horng R-H, Lin C-Y, and Wuu D-S 2015 ACS Appl. Mater. Interfaces 7 (4), pp 2546–2553.
[6] Chou, T.P., et al. 2008 J. Nanophotonics 2 pp. 023511.
[7] Purwanto A, Widiyandari H, and Jumari A 2012 Thin Solid Films 520 (6), pp. 2092–2095.
[8] Russo B, and Cao G.Z 2008 Appl. Phys. A 90, pp. 311–315.
[9] Suyitno, Arifin Z, Santoso A.A, Setyaji A.T, and Ubaidillah 2014 Appl. Mech. Mat. 575, pp 689-695.
[10] Senthilkumar V, Vickraman P, and Ravikumar R 2010 J. Sol-Gel Sci. Tech. 53(2) pp 316–321.
[11] Shi, X.H. and K.J. Xu 2017 Mat. Sci. Semiconductor Process. 58, pp. 1–7.
[12] Chavarria-Castillo K.A et. al. 2016 Mat. Research 19 pp. 97-102.
[13] Banyamin Z.Y, Kelly PJ, West G, and Boardman J 2014 Coatings 4, pp. 732-746.
[14] Montero, Chialvo, M.R.G and Chialvo, A.C 2009 J. Mater. Chem. 19 3276–3280.
[15] Supriyono, Surahmana H, Krisnandia Y.K, and Gunlazuardia 2015 J. Procedia Environ. Sci. 28 pp. 242–251
[16] Saehana S, Darsikin, Yuliza E, Khairurrijal and Abdulah M 2012 J. Sol. Energy Eng. 136, pp. 044504-1.
[17] Saehana S, Muslimin, and Abdulah M 2014 J. Renew. Sustain. Energy. 6, pp. 023109.
[18] Saehana S, Arifin A, Khairurrijal, Abdulah M 2012 J. Appl. Phys. 111, 123109.

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