Growth of TiO_2-ZrO_2 Binary Oxide Electrode for Dye Sensitized Solar Cell Application

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Abstract **– TiO_2-ZrO_2 fine binary oxide was prepared by mechanochemical milling process to be homogeneous binary oxide powder. TiO_2-ZrO_2 paste was deposited on microscopic glass slide by rolling. It was immersed in the henna solution and annealed at 100°C for 2h. It was deposited onto another glass slide and used as counter electrode (second electrode). Two glass slides were offset and two binder clips were used to hold the electrodes together. Photovoltaic properties of TiO_2-ZrO_2 cell were measured and it was expected to utilize the dye sensitized solar cells application.**

Keywords**— mechanochemical milling, carbon catalyst, photovoltaic properties**

I. INTRODUCTION

Dye sensitized solar cells (DSSCs) based on nanocrystalline inorganic oxides such as TiO_2, ZnO and SnO_2 have attracted much attention since their first description in the beginning of 1990s by Gratzel and O'Reagan [1]. DSSCs, also known as Gratzel cell, are new inventions in thin film solar cells. The DSSCs are currently attracting extensive academic and industrial interest envisioning this technology as a powerful and promising way to generate electricity from the sun [1]. The DSSCs convert the energy in light absorbed by dyes or pigments into other forms of energy. The DSSCs realized the optical absorption and the charge separation process by the association of a sensitizer as light-absorbing material with a wide band gap semiconductor of nano-crystalline morphology.

Titanium dioxide, also known as titanium (IV)oxide or titania, is the naturally occurring oxide of titanium, chemical formula TiO_2. TiO_2 has proved to be one of the most promising materials for various applications such as solar energy conversion, fuel cells, paints and photocatalysts, due to its high chemical stability, availability and low cost [2]. TiO_2 is a wide band gap oxide. TiO_2 nanoparticle has been used as the photoelectrode in DSSCs. Because of its high specific surface area that allows the absorption of a large number of dye molecules [1]. In DSSCs, TiO_2 only absorbs UV and does not absorb other wavelengths. Therefore, the energy conversion efficiency is increased by adding a dye that absorbs light with wavelengths in the visible light range of the solar spectrum. Zriconium dioxide (ZrO_2) have been investigated for their catalytic properties with organic compounds [2]. ZrO_2 has been used not only as a support for TiO_2 but also with TiO_2 as a binary oxide catalyst since ZrO_2 itself can act as a photocatalyst [3-4]. A mixture of ZrO_2 and TiO_2 was used as a electrode for dye- sensitized solar cell. Not only chemical dye but also natural dye can be used in DSSCs.

Dye Sensitizer

Henna leave was used in this work.

Henna plant profile

Scientific Name	- Lawsonia inermis L.
Myanmar Name	- Dan
English Name	- Henna plant
Family	- Lythraceae

II. PREPARATION OF MIXED BINARY OXIDE TiO_2-ZrO_2

Titanium dioxide (TiO_2) (BDH chemicals) was used in this work. The TiO_2 was prepared by using a mortar and pestle, mesh-sieving and ball-milling method. Firstly, the TiO_2 powder was put in a mortar and pestle. Then, it was ground for 1h to reduce particle size, as shown in Figure 1.

Figure 1. A mortor and pestle

Next, the TiO_2 was also mesh sieved with 3 step mesh to get uniform and the lightest particles. One set of mesh-sieves was described at Figure 2.

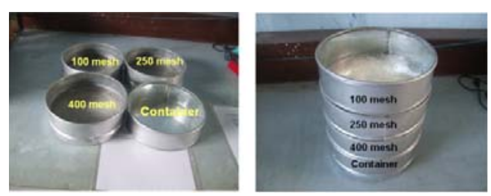

Figure 2. Three-stage mesh sieves

After sieving, the TiO_2 was milled with the ball-milling machine for 20h to reduce the particle grain size and shown in Figure 3.

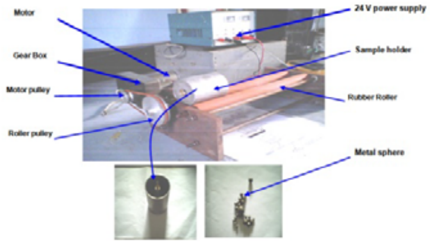

Figure 3. Ball-milling machine

After doing this state, the 9 ml of ethanol (C_2H_5OH) were added into the TiO_2 powder as a binding agent. This powder was continuously stirred by a magnetic stirrer for 2h to be homogeneous. It is shown in Figure 4.

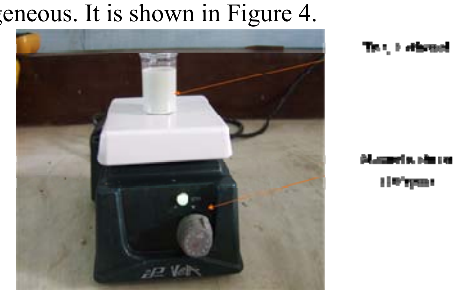

Figure 4. Magnetic stirrer

Then, it was dried at room temperature for 24h. Eventually, the homogeneous TiO_2 was formed. Similarly, the next sample of zirconium dioxide (ZrO_2) (AnalaR-grade) was made. Eventually, the homogeneous ZrO_2 was found. Then, TiO_2 (95%mol) and ZrO_2 (5%mol) were mixed together. The 9 ml of ethanol (C_2H_5OH) were added into the TiO_2- ZrO_2 powder as a binding agent and stirred with magnetic-stirrer for 2h. The mixture powder was annealed at 450°C, 500°C, 600°C and 700°C for 1h in oxygen ambient. Crystal structure of mixed binary oxide was analyzed by X-ray diffraction (XRD) technique.

TiO_2-ZrO_2 binary oxide characterization

XRD is a non-destructive technique for the qualitative and quantative analysis of the crystalline materials, in form of powder or solid. The information about the crystallographic properties such as crystallite size and lattice parameters of all samples has been obtained from the XRD analysis. The TiO_2-ZrO_2 binary oxide was annealed at 450°C, 500°C, 600°C and 700°C using XRD technique. They were shown in Figure 5 (a-d)

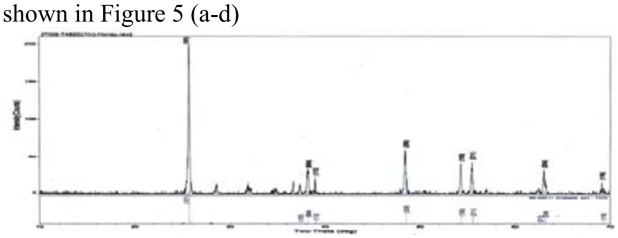

Figure 5 (a) XRD pattern of TiO_2-ZrO_2 binary oxide at 450°C

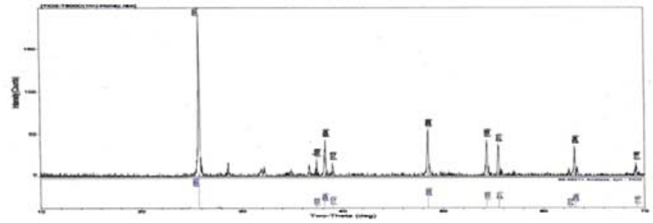

Figure 5 (b) XRD pattern of TiO_2-ZrO_2 binary oxide at 500°C

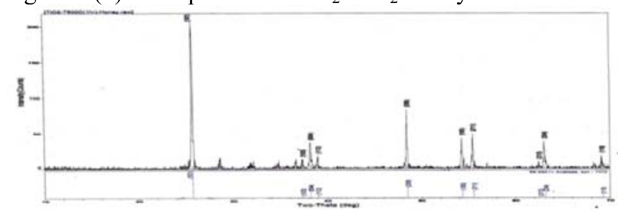

Figure 5 (c) XRD pattern of TiO_2 – ZrO_2 binary oxide at 600°C

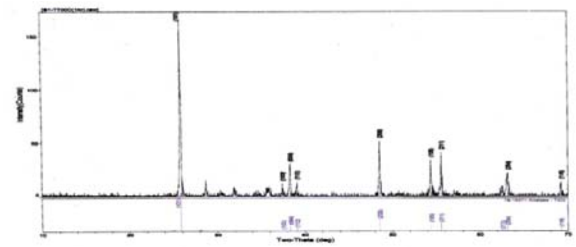

Figure 5(d) XRD pattern of TiO_2-ZrO_2 binary oxide at 700°C

On the XRD patterns, ten peaks were clearly observed. This peaks were compared the data from the JCPDS library file. The most dominant peak was occurred at (101) peak. The lattice distortion (or) lattice strain of other peaks after annealing indicates the good crystalline nature. Among the temperature, 600°C is the best.

TiO_2-ZrO_2 paste preparation

Six grams of TiO_2-ZrO_2 binary oxide were put in a mortar and pestle. Next, the appropriate amount of distilled water and dish washing detergent were added in a mortar and pestle. Then, 9ml of nitric acid were sucked with large eyedropper and drop by drop into a mortar and pestle. After grinding 30 minutes, TiO_2-ZrO_2 paste will be formed. They were shown in figure 7(a and b)

Figure 7(a) Nitric acid, TiO_2-ZrO_2, distilled water and dish washing detergent

Figure 7(b) TiO_2-ZrO_2 paste

III. PREPARATION OF DYE SENSITIZER

Henna leaves were used in this work. Firstly, henna leaves were ground in a mortar and pestle and squeezed them. The green solution of henna was found. They were shown in figure 8 (a and b).

Figure 8(a). Henna leaves mortar

Figure 8(b) Green solution of henna (after grinding)

Then, the green solution of henna was sieved with the filter paper. After sieving 1h, this solution was turned to red solution. The red solution boiled with hot plate at 100°C. Then, it was cooled and sieved them 3 times with the filter paper. The red colour of solution was found. It was shown in Figure 8 (c and d).

Figure 8(c) Sieved this solution 3 times with the filter paper

Figure8 (d) Red solution of henna

After doing this state, the 40ml of henna solution and 4 ml of ethanol were mixed together. Then, they were annealed for 2h with heat controller at 100°C. After cooling at a moment, they were sieved with the filter paper. Eventually, the henna solution was retained and it was used in DSSCs. They are shown in Figure 8 (e-g).

Figure 8(e) Henna solution annealed with heat controller

Figure 8(f) Filtered the solution paper

Figure 8 (g) Henna solution with the filter paper

Preparation of carbon catalyst

Soot was used as carbon catalyst counter electrode. Firstly, 11.64g of soot were put in a mortar and pestle. Then, 3ml of hydrochloric acid (HCL) were sucked with a large eye dropper and drop by drop into a mortar and pestle. After grinding 1h, the carbon solution was retained and then it was used as a catalyst. They were shown in Figure 9 (a-c).

Figure 9(a) Soot

Figure 9(b) Hydrochloric acid (HCL) and soot

Figure 9(c) Mixture of HCl and soot

IV. PREPARATION OF DSSC

A. Preparation of microscopic glass

Microscopic glasses were used in this work. Firstly, 2 x 2 cm square microscopic glasses were washed in the solution of soap for 5 min. And then, it washed with acetone for 5 min. Then, they were dried at room temperature for 15min. It was shown in Figure 10(a).

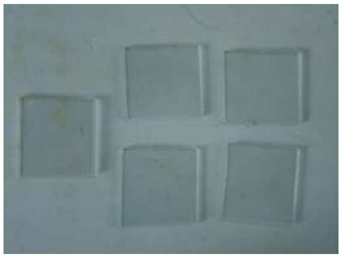

Figure 10(a) Dried microscopic glass

The LCR meter was used to identify the conductivity of two surfaces of glass slide. It was marked the side of good conductivity. After doing this state, in both side of glass were covered with masking tape. It is shown in Figure 10(b)

Figure 10(b) Preparation of glass with masking tape

The glass plates are checked using a LCR meter to make sure that the conducting side is facing up, and then TiO_2 paste is applied to conducting side of the glass using a roller and quick downward sweeping motions, as shown in Figure 11.

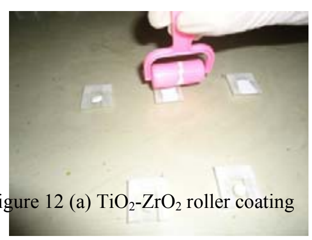

Figure 12 (a) TiO_2-ZrO_2 roller coating

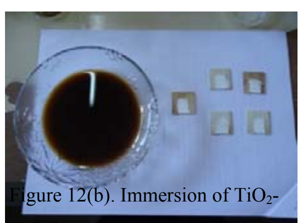

Figure 12(b). Immersion of TiO_2-ZrO_2 layer in dye sensitizer

In order to be dry and strengthen the TiO_2 coating, the plates were heated over an oven for 1 h. Once they have cooled, they were placed in a dish filled with sensitizing dye for 1h and they were annealed for 1h with heat controller at 200° C. They were shown in Figure 12 (a and b).

B. Preparation of the positive counter electrode

The soot were sprayed on the glass with substrate temperature at 200^{o}C and shown in Fig 13(a). Application of the carbon spray was accomplished by taping off the edges of the glass plate.

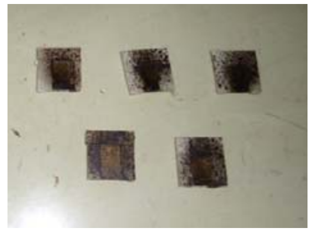

Figure 13 (a) Carbon catalyst on glass slide

Figure 13(b) Carbon electric with Iodine

Preparation of the positive and negative electrodes is completed, 1-2 drops of mediator were placed on the negative electrode and shown in Figure 13 (b). Two prepared glass slides were set together and the sandwiching of the two plates was offset so that each one had a small position exposed so that an alligator clamp could be attached and indicated in Figure 14.

Figure 14. DSSCs with binder clips

V. SOLAR CELL EVALUATION (UNDER SUNLIGHT)

Figure 15 showed the change in photocurrent as a function of voltage with henna dye sensitizer at different PH levels. Some solar cell parametres such as conversion efficiency and fill factor were evaluated and listed in Table 1(a-b).

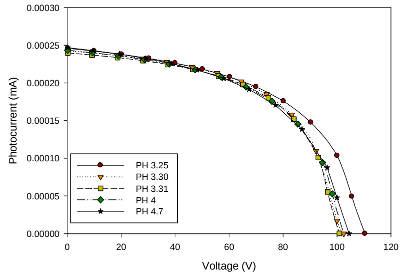

Figure 15 Current-voltage curves for Henna sensitized solar cell

TABLE

I(A) SOLAR CELL PARAMETERS OF THE CELLS WITH NATURAL DYE AT DIFFERENT PH LEVEL

PH Level	I_m (mA)	V_m(mV)	I_{sc} (mA)	V_{oc}(mV)
3.25	1.86e-4	75.00	2.47e-4	110.26
3.30	1.84e-4	73.51	2.43e-4	100.75
3.31	1.88e-4	69.51	2.40e-4	100.75
4.00	1.95e-4	66.25	2.45e-4	102.48
4.70	1.88e-4	69.00	2.47e-4	104.02

TABLE

I(B) EFFICIENCY AND FILL FACTOR OF THE CELLS WITH NATURAL DYE AT DIFFERENT PH LEVEL

PH Level	Efficiency (%)	Fill Factor
3.25	0.088	0.511
3.30	0.085	0.553
3.31	0.083	0.541
4.00	0.088	0.505
4.70	0.082	0.511

VI. CONCLUSION

Growth of TiO_2- ZrO_2 binary oxide electrodes DSSC with henna leave sensitizers and their solar cell properties have been successfully implemented. From the experimental results, it was found that the conversion efficiency was slightly decreased with an increase in PH level of dye sensitizer. The fill factors of all fabricated cells were observed to be about 0.5 and indicated the industrial realization. Thus it definitely meets the special requirements for DSSC of low cost and Eco-friendly.

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