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Synthesis of Nanopartical-based Binary Oxide Electrode TiO_2-ZrO_2 with Carrot-derived Natural Dye Extract for Dye Sensitized Solar Cell (DSSC) Application

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ABSTRACT

Dye-sensitized solar cell (DSSC) is the third generation of thin film solar cell. In this work, carrot fruit dye was prepared and used in DSSC as a sensitizer. TiO_2–ZrO_2 fine binary oxide was mechanochemically prepared and made paste. TiO_2–ZrO_2 paste (colloidal) was deposited onto FTO/glass in two ways, i.e. as single and double coatings by rolling method. It was immersed in the carrot solution to get dyed cell. The dyed TiO_2-ZrO_2-FTO/glass cell was offset with the FTO/glass cell coated with adhesive carbon paste. Photovoltaic properties of prepared TiO_2–ZrO_2 DSSC cell were measured. Results showed that the efficiency of double coating cell was greater than that of the single coating cell. According to the fill factor from the experiments, both the DSSC cells were acceptable for industrial requirement. Hence, it can be concluded that the results obtained were acceptable in the use of cost-effective and eco-favourable dye-sensitized solar cell.

Keywords: Mechnochemical milling, FTO/glass, carbon counter electrode, photovoltaic properties dye-sensitized solar cells

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INTRODUCTION

Photoelectrochemical application based on dye-sensitized nanostructured TiO_2 solar cell has attracted much attention as a low cost alternative to conventional silicon solar cell. In 1991, dye-sensitized solar cell (DSSC) based on liquid electrolyte with an efficiency of about 11% was first reported by O' Regan and Gratzel (Buradah *et al*., 2011;

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Nupearachchi *et al*., 2011; Zhou *et al*., 2011). DSSCs, which are also known as Gratzel cells, are new invention in thin film solar cell. The DSSCs are currently attracting extensive academic and industrial interests envisioning this technology as a powerful and promising way to generate electricity from the sun (Flores *et al*., 2007; Fujihara *et al*., 2007; Ali & Nayan, 2010; Kakuta *et al*., 2012; Ekanayake *et al*., 2011).

The main principal use of these DSSCs is the dye-sensitization of wide band gap semiconductors such as TiO_2, ZnO, SnO_2, etc., by suitable regenerative dyes which ultimately convert solar irradiation into electricity (Win *et al*., 2012). TiO_2 is a wide band gap oxide and it has been used as photoelectrode in DSSCs because of its high specific surface area that allows absorption of a large number of dye molecules. TiO_2 has been proven to be one of the most promising materials for various applications including solar energy conversion, fuel cells, paints and photocatalysts, due to its high chemical stability, availability and low cost. 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 (Neppolian *et al*., 2007; Mane *et al*., 2005; Gratzel, 2004; Gratzel, 2003; Kim *et al*., 2005; Park *et al*., 2003; Win *et al*., 2012). ZrO_2 has been investigated for its catalytic properties with organic compounds. In addition, 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 as a photocatalyst (Wongcharee *et al*., 2006). The ability of sensitizers in the natural dye is linked to Anthocyanin properties. Anthocyanin molecules in the forms of carbonyl and hydroxyl which occur naturally in fruit, leaf and flowers are responsible to show types and colour pigments in visible red-to-blue spectrum. Natural dyes have wider absorption spectra compared to chemical synthesis dyes due to the more various constituents in the natural dyes (Lin *et al*., 2007; Okoli *et al*., 2011; Abeygunawardhana *et al*., 2011). In DSSCs, natural dyes can help to expand their absorption spectra. In this work, carrot solution was used as a natural dye sensitizer.

MATERIALS AND METHOD

Preparation of Mixed Binary Oxide TiO_2-ZrO_2

Titanium dioxide (TiO_2) (BDH Chemical) was used in this work. The TiO_2 nano particle was prepared by using a motor and a pestle, mesh-sieving and ball-milling method. Firstly, the TiO_2 powder was ground for 1h before sieving it with 3-step meshes and milling it with the ball-milling machine for 20h to get nano particle grain size. Meanwhile, 9 ml of ethanol (C_2H_5OH) was added into the TiO_2 powder as a binding agent. This powder was continuously stirred by using a magnetic stirrer for 2h to be homogeneous. Then, it was dried at room temperature for 24h. Eventually, the homogeneous TiO_2 powder was obtained. Zirconium dioxide (ZrO_2) (AnalaR-grade) was used in the preparation of the homogeneous ZrO_2 powder following the same procedure as TiO_2. Then, TiO_2 (95%mol) and ZrO_2 (5%mol) were mixed thoroughly in 9ml of ethanol (C_2H_5OH) and stirred for 2h. The mixture powder was annealed at four different temperatures of 450ºC, 500ºC, 600ºC and 700ºC for 1h in oxygen ambient. Crystal structure of mixed binary oxide TiO_2 –ZrO_2 was then analyzed by using X-ray diffraction (XRD), as shown in Fig.5.

TiO_2-ZrO_2 Paste Preparation

TiO_2-ZrO_2 powder was dissolved in acetylacetone and water mixed solution. During this preparation, detergent was added into it as a surfactant. It was coated onto clean FTO/glass substrate using the rolling method so as to obtain the single layer cell and double layer cell. After air drying, it was sintered at 400ºC for 1 h. Both films were immersed in 0.1 M HCL for 1 h and washed with DIW maintained 1 h for surface treatment. Finally, TiO_2-ZrO_2 film (active area = 1cm × 0.5cm) was formed on FTO/glass substrate.

Preparation of Dye Sensitizer

Commercial Carrrot (Daucus carota L.) was used as a dye. Firstly, the carrot sticks were cured in boiled water for 45 min to attain softer and enhanced colour. The outside layer of the carrot skin was peeled off and sliced into several pieces. Secondly, the pieces of carrot was packed in a piece of thin cloth and cured in ehanol. Ethanol changed its colour into orange. After cooling, pH was measured to be 6. Later, some powder was acidified with 1% H_2SO_4, mixed with ethanol and annealed at 80ºC for 1 h. After cooling, the pH level was found to be 8. The solutions obtained from two different methods were mixed and pH9 was obtained. The KOH base was added to get the natural carrot solution. Fig.1 shows the carrot pieces and carrot solution.

Fig.1. Carrot pieces and carrot solution

Preparation of Carbon Catalyst

First of all, KOH (16 ml) and ethanol (8 ml) were mixed together. Then, carbon powder and black carbon powder were dispersed into this mixture solution. After dispersion, carboxylmethylcellulose (0.24 g) was also added and adhesive carbon paste was formed. It was coated onto FTO/glass substrate (active area = 1cm × 0.5cm) by rolling and annealing it at 180ºC for 1 h.

Preparation of FTO/glass

FTO powder was prepared by the solution method. Firstly, tin (IV) chloride (10.5g) was added to 150ml ethanol and mixed in a sealed glass vial for 5h. In another sealed glass vial, ammonium fluoride (1.86g) was added into DIW (5.04ml) and mixed for 5h. The fluoride solution was added into the glass vial containing the tin chloride solution while in a water bath at 60ºC temperature. Finally, the FTO powder was obtained. Then, it was coated onto the glass by spinning method.

DSSC Prototype Preparation

The TiO_2-ZrO_2 coated electrodes were immersed in the carrot solution for 15 h and annealed at 100ºC for 1 h. Fig.2 shows the carbon coated and TiO_2-ZrO2 coated electrodes.

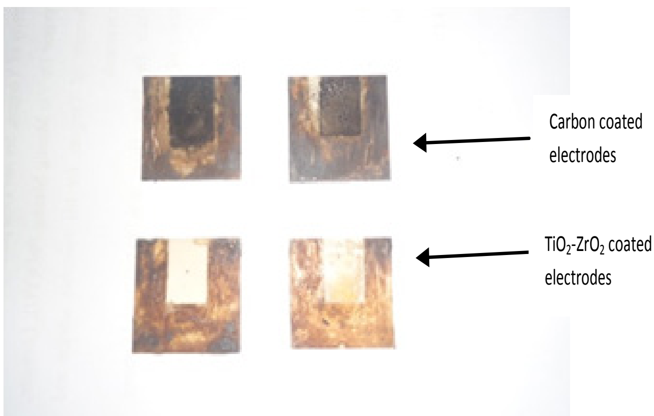

Fig.2: Carbon coated and TiO_2-ZrO_2 coated electrodes

When the preparation of the positive and negative electrodes was completed, 1-2 drops of iodine were placed on the negative electrode. 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 as indicated in Fig.3.

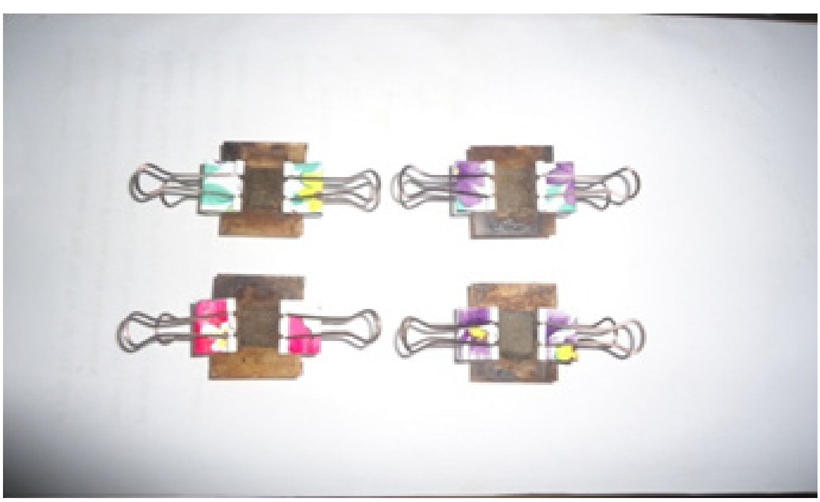

Fig.3: DSSCs with binder clips

RESULTS AND DISCUSSIONS

Characterization of TiO_2-ZrO_2 Binary Oxide Power

On the XRD pattern of TiO2-ZrO2 binary oxide power ten peaks were clearly observed. The most dominant peak occurred at (101) peak shown TiO_2 Anatase structure. The lattice distortion (or) lattice strain of other peaks after annealing indicates the good crystalline nature. Meanwhile, 600ºC is shown to be the best temperature for this purpose. Fig.4 shows the XRD pattern of TiO_2-ZrO_2 binary oxide at 600ºC.

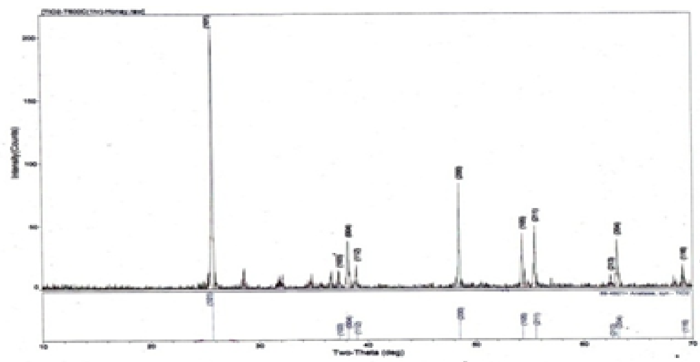

Fig.4: The XRD pattern of TiO_2-ZrO_2 binary oxide at 600ºC

Solar Cell Evolution

Fig.5 shows the change in photocurrent as a function of voltage with carrot solution. Some solar cell parameters such as conversion efficiency and fill factor were evaluated and listed in Table 1 and Table 2.

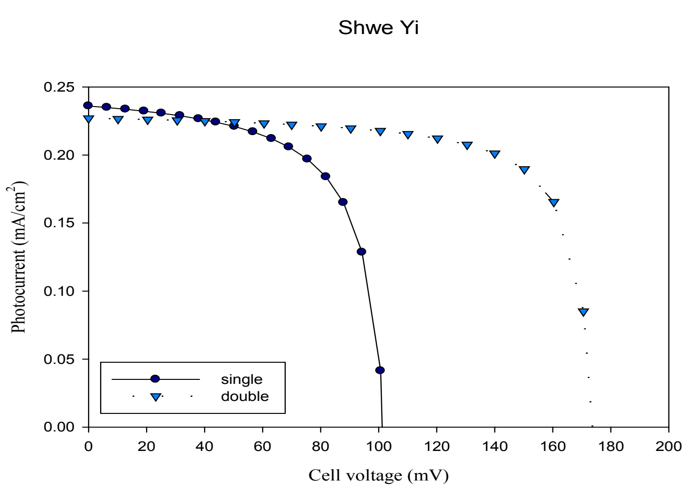

Fig.5: Current-voltage curves for Carrot sensitized solar cell

TABLE 1: Solar cell parameters of the cells with natural dye

TiO_2-ZrO_2	Im (mA)	Vm (mV)	Isc (mA)	Voc (mV)
Single	29.3	75.4	35.4	10.3
Double	28.6	150.0	34.1	17.4

TABLE 2: Efficiency and fill factor of the cells with natural dye

TiO_2-ZrO_2	Efficiency (%)	Fill Factor
Single	0.015	0.61
Double	0.030	0.72

CONCLUSION

Preparation of TiO_2-ZrO_2 binary oxide electrode (single & double layer coatings) with carrot extract was implemented in this study. The conversion efficiency of TiO_2-ZrO_2 (double coating) (0.030%) was found to be higher than that of binary oxide at single coating (0.015%). The increase in efficiency was found to be due to the film thickness of coating. The fill factor of both the DSSC films was found to be within the range of accepted value for industrial purposes.

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