

STUDY ON CONSISTENCY AND DENSITY OF TITANIUM DIOXIDE AND TUNGSTEN TRIOXIDE COATINGS ON CONDUCTIVE GLASS FROM DIPPING PROCESS AFFECTING LIGHT ABSORPTION AND TRANSMISSION

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Abstract

Light property is a crucial factor in photovoltaic power generation. Therefore, the study of light absorption and transmission of materials is important for developing of solar cell coating material. Titanium dioxide (TiO₂) and tungsten trioxide (WO₃) are the important candidates for use as the solar cell coating material. Because TiO₂ and WO₃ have a narrow energy band gap and have photocatalytic properties which are the essential properties for the solar cell application. The aim of this work is to investigate the light absorbance ability of the TiO₂ and WO₃ coating layer. The gel of TiO₂ and WO₃ was synthesized by the sol-gel method. The synthesized gel of TiO₂ and WO₃ was coated on the conductive glass (FTO) by using the dip-coating technique. The light absorbance of the coated surface was examined by the UV-Visible Spectrophotometer technique (UV-Vis). The surface of coating homogeneity and the coating density were evaluated by 3D imaging which was investigated by 3D reconstruction of the samples by Synchrotron X-ray tomographic microscopy (SR-XTM) technique. The results show the light absorption ability of the conductive glass coated by WO₃ is higher than that of TiO₂. However, the coating homogeneity and the coating density of the WO₃ coated layer are lower than that of the TiO₂ coated layer. The unevenness of the WO₃ coated layer is the cause of non-uniform light absorption. The homogeneity of the coating layer is decreased with increasing the coating layer number because the coating is unevenly distributed resulting in some areas are denser than others. From all the experimental results, it is concluded that even the TiO₂ coated layer has a tendency to increase the absorbance less than WO₃ coated layer, but it is acceptable considering of uniformity coated layer. Consequently, TiO₂ is suitable for applying to solar cells because of its increasing of absorbance capacity and reducing of light reflection.

Keywords: Solar cell; Photocatalysis; Photovoltaic application; Synchrotron X-ray tomographic microscopy

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Introduction

Alternative energy is increasingly being used to generate electricity these days. The most well-liked is the production of electricity using solar power. The idea behind solar energy production is the conversion of light energy into electrical energy using photovoltaic cells. The solar cells' construction material has an impact on how efficiently photovoltaic energy is produced. The material is suited for use in photovoltaic panels due to its strong light absorption capabilities and low energy band gap. As a result, while developing new solar cell materials, absorbance properties and energy band gap are taken into consideration. Polycrystalline silicon is the substance with the aforementioned qualities that is now most frequently used to produce solar cells. Even though polycrystalline silicon has a narrow energy band gap and strong optical absorption, there are still constraints on how much power can be produced, regardless of lifetime, the absorbance range's limitation and other factors that influence the effectiveness of electricity generation. Therefore, scientists focus on developing materials that enhance solar cells' capacity to generate power. The most commonly used materials are photocatalysis materials with good light absorption capability. The solar cells' ability to absorb energy is increased as a result. More energy can be generated by the solar cell as the absorbance rises. Materials such as CaS, CaTe, TiO₂, WO₃ and etc. are most commonly utilized. 90% of the sunlight is absorbed by CaS and CaTe, demonstrating their excellent light-absorbing abilities (Guo *et al.*, 2015; Shankar *et al.*, 2017). However, Substances containing cadmium were later discovered to be harmful to the environment. Currently, it is not well-liked. For producing solar cells, TiO₂ and WO₃ are widely employed. These materials are coated using a variety of processes, including silk screening, spin coating, dip coating and etc (You *et al.*, 2019).

The multifaceted approach to the practical applications of photocatalysis while taking into account different technical and material aspects (Emin *et al.*, 2015). Which consists of hybridization of photocatalysis with other technologies, photocatalytic reactor development (Shen *et al.*, 2018), photocatalyst immobilization methods, low-cost photocatalytic materials, photocatalytic activities as a function of the wavelength and durability of photocatalysts against deactivation and fouling (Satyapaul Singh and Giridhar, 2013; Weon, 2018; He *et al.*, 2021). However, surface morphology, agglomerated size, thickness, density, homogeneity or consistency of the coated layer,

among other factors, play a significant role in determining the parameters of the photocatalytic process (Pelizzetti *et al.*, 1990; Kumar and Pandey, 2017). Currently, there are many technologies and techniques to detect such a factor. One of the techniques that has gained attention and which builds credibility in material inspection and analysis is synchrotron light technology.

XTM beamline located at BL1.2W is the experimental facility for X-ray tomographic microscopy. With the synchrotron radiation generated from the 2.2-Tesla multipole wiggler of Siam Photon Source (SPS), the XTM beamline offers the high-intensity x-ray beam for microtomography to provide the reconstructed cross-sectional detail and 3D visualization of various kinds of samples at the resolution up to 1 micron (0.72-micron pixel size). The application of X-ray tomographic imaging is appropriate for research projects that seek to describe things in three dimensions, such as the internal morphology of samples, engineering defects, porosity, surface roughness, and pore distribution (Phakkhananan *et al.*, 2021; Utchawadee *et al.*, 2021; Kantrong *et al.*, 2022).

This research aims to investigate the uniformity and density of titanium dioxide and tungsten trioxide coated layers using Synchrotron radiation X-ray tomographic microscopy (SRXTM) technique coating on conductive glass by a dipping procedure that influences the ability of absorbance and transmittance. The titanium dioxide and tungsten trioxide materials were first synthesized by using the sol-gel method. Prior to analyzing the fundamental characteristics, such as structural phase, absorbance, and transmittance properties using XRD and UV-Vis, respectively. The results were then confirmed and further supported by science using the SRXTM method on how the coating layer's density and homogeneity affect optical properties.

Materials and Methods

Synthesis of Titanium dioxide (TiO₂) and Tungsten trioxide (WO₃)

Synthesis of Titanium dioxide by Sol-gel method

Titanium (IV) isopropoxide (TTIP) (ACROS ORGANICE, 98%) was used as a precursor for preparation of Titanium dioxide (TiO₂). Titanium (IV) isopropoxide was first dissolved in ethanol and

then stirred with 600 - 800 rpm for 15 mins. After stirring for 15 min, the reactants begin to react and form to be a clear white solution. Hydrochloric acid (HCl) with concentration of 2 molar (M) was applied in a clear white solution to adjust the pH to a value of 3. Continue stirring the clear white solution with the same speed but increase the spinning time to 30 mins, The white solution is gradually reacted with HCl and turns into a milky white gel of TiO_2 .

Synthesis of Tungsten trioxide by Sol-gel method

Sodium tungstate (Na_2WO_4) (LOBA, 99%) was used as a starting material and then first dissolved in deionized water (DI) to form a clear white solution. During stirring with speed of 600 - 800 rpm for 15 mins, the clear white solution was acidified with 20 ml of nitric acid (HNO_3). For this stage, slowly drop of HCl into the solution, it can be seen that the solution gradually reacted and color change to yellow. Then continues stirring for 15 mins with the same speed, to make the chemical completely reacted until a yellow-green solution is obtained. Once the impurities were filtered and washed with deionized water more than 10 times, to achieve a purity phase of yellow wet powder. Final step, yellow wet powder and 2 ml of hydrogen peroxide (H_2O_2) were mixed and stirred with 600-800 rpm for 2 hrs to get a yellow clear gel of WO_3 .

Characterization of Titanium dioxide and Tungsten trioxide coated layers

Preparation of Fluorine doped Tin Oxide (FTO) glass specimens

Fluorine doped Tin Oxide (FTO) was used as a glass substrate surface. Before using, FTO glass substrates were cut into 2 cm $\times$ 3 cm rectangle before being washed to reduce the contamination on the surface before use. First sequence for washing is used ultrasonic cleaner with deionized water at 60°C for 15 mins. Then, first cleaned FTO was washed with ethanol again by using an ultrasonic cleaner at room temperature for another 15 mins. The washed FTO Workpieces until finished all the steps will be dried and stored in a desiccator cabinet to be ready for use.

Preparation of TiO_2 and WO_3 coated on FTO conductive glass

TiO_2 and WO_3 gel were successfully coated on FTO conductive glass substrates by using typical dip coating technique. The number of TiO_2 and WO_3 coated layers is the main factor involved in the study on the effect of light absorption and

transmission. Before testing, all coated sample were heat treatment at a temperature range of 450 and 500°C for 2 hrs to achieve a single phase of TiO_2 and WO_3 coated on FTO conductive glass substrates. FTO/ TiO_2 and FTO/ WO_3 samples are now led to the next step of characterization and properties test.

Structural phase analysis of TiO_2 and WO_3 coated material

The structural phase of FTO/ TiO_2 and FTO/ WO_3 samples was identified by X-ray diffraction (XRD, Bruker D2 PHASER) with the analytical 2-theta range of 10°-70°, using a step time of 0.5 sec/step and step size of 0.02°/step.

Surface analysis of TiO_2 and WO_3 coated layer on FTO glass

The surface of FTO, FTO/ TiO_2 and FTO/ WO_3 samples was identified by Field Emission Scanning Electron Microscope (FE-SEM, model MIRA3) with the analytical magnification 100 x and 5.00 kx.

Synchrotron radiation X-ray tomographic microscopy

X-rays were carried out as the spectral range of synchrotron radiation for computing 3D tomographic reconstruction at 1.2 W beamline [Synchrotron Light Research Institute (SLRI), Thailand]. X-ray radiation changed the amplitude of a wave after going through the sample. When maximum height of the amplitude was low after passing through the sample, X-ray radiations were absorbed in heavy or high attenuation materials, which caused a dark shadow in the radiographic image. On the other hand, if the height of the amplitude was still high after passing through the sample, most of the X-ray radiations were transmitted, which caused a bright shadow in the radiographic image. As a result, one radiographic image can have many shades of shadow depending on the attenuation of the materials in the sample. The radiographic images of the sample were collected at 180° with 0.1 angular increments for a complete dataset. In order to minimize the artifacts, a polychromatic X-ray was attenuated with 200 micron-thick aluminum foil, with a mean energy of about 11.5 keV. The radiographic images were collected on a sCMOS camera with a pixel size of 1.44 μm . The data was pre-processed and reconstructed in three dimensions based on a filtered-back projection algorithm using Octopus Reconstruction software. After that, reconstructed images were visualized by using Drishti software. An example for analytical procedure with the SRXTM technique.

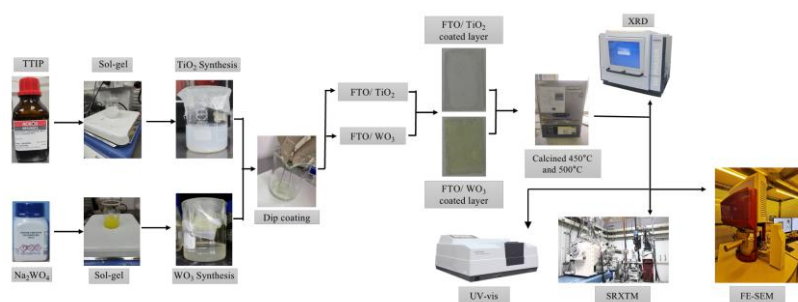


Figure 1. The overall flow chart of TiO₂ and WO₃ preparation and characterizations

Optical Properties of FTO/TiO₂ and FTO/WO₃ samples

Cary 300 UV-Vis spectrophotometer was used to analytical the light absorption and transmission of FTO glass, FTO/TiO₂ and FTO/WO₃ coated material. Absorption and transmission are measured in the wavelength range of 200-800 nm as this is because of a comprehensive range for using materials on solar cells.

The overall flow chart of material preparation, method and characterizations of titanium dioxide and tungsten trioxide coated layers was shown in the Figure 1.

Results and Discussion

Structural phase analysis of TiO₂ and WO₃ coated material

The structural phase of TiO₂ and WO₃ coated materials synthesized via sol-gel method was characterized by X-ray diffraction. As the XRD result from Figure 2(a), TiO₂ coated material after heat treatment at 450°C and 500°C exhibited sharp diffraction peaks attributed to mixed-phase of TiO₂ anatase phase and TiO₂ rutile phase. The single phase of TiO₂ anatase phase and TiO₂ rutile phase can be confirmed by JCPDS no. 21-1272 and JCPDS no. 21-1276, respectively. In the case of WO₃ coated material after heat treatment at 450°C, the imperfect phase or second phase of WO₃·XH₂O (JCPDS no. 35-1001) is still found and mixed with the complete phase or single phase of WO₃ (JCPDS no. 83-0948). However, when heat treatment at 500°C, WO₃·XH₂O was completely reacted with temperature and evaporated the chemical water, then finally form to be completely single phase of WO₃. From the XRD results, it is revealed that the heat treatment temperature of TiO₂ and WO₃ coated material is not the same firing condition. The temperature used to form the single phase of TiO₂ is wider than the temperature used to form a single phase of WO₃.

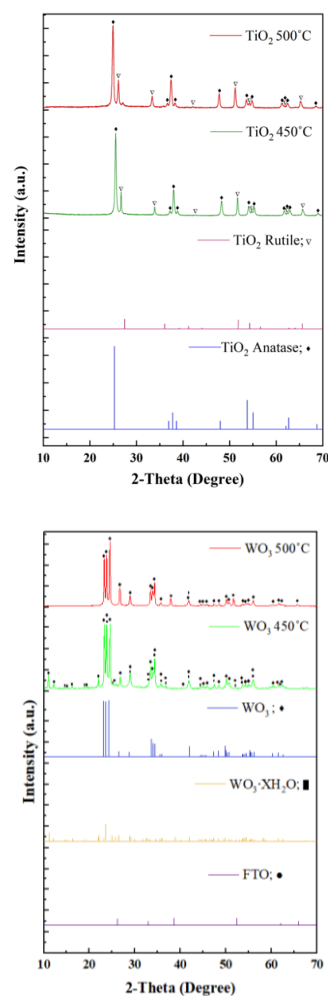


Figure 2. XRD pattern of TiO₂ and WO₃ after heat treatment at 450°C and 500°C

Figure 3 shows the surface morphology of (a.) FTO glass, (b.) FTO/TiO₂ and (c.) FTO/WO₃ by FE-SEM technique. It was observed that TiO₂ exhibits a more uniform coated dispersion over the FTO-substrate than WO₃. Additionally, it was found that the particles of TiO₂ were smaller than

the particles of WO_3 coated on the workpiece surface, suggesting that TiO_2 produced using the sol-gel method is more reactive of chemical reaction than WO_3 . In addition, fissures can be seen all over the FTO-surface when looking at WO_3 coated layer. The surface tension of the fairly liquid WO_3 sol-gel is really what makes the WO_3 coatings clump and produce unequal distribution on FTO-substrate. The FE-SEM technology was used to examine the coating layer's surface that the unevenness of the coated layer not only influences the density of the coated layer but it also effected to the optical properties of the material.

From Figure 4, the 3D reconstruction with gradient color from blue to red was applied to identify the density of materials. It was found that

the pure FTO glass (Figure 4(a)) showed the blue color due to a low density. Whereas the FTO glass with TiO_2 (Figure 4(b-d)) showed the green color. From this result, it implies that TiO_2 has a higher density than the pure FTO glass. It also found that TiO_2 particles were homogeneously coated on the FTO glass. After dipping more layers of TiO_2 particles on the FTO glass, the density was higher by changing from light green to dark green color as shown in Figure 4(b-d).

Nevertheless, after applying the dipping process for WO_3 particles, the 3D reconstruction showed the blue color as shown in Figure 4(d-f). That means WO_3 particles were barely coated on the FTO glass except the edge of the sample (red arrow).

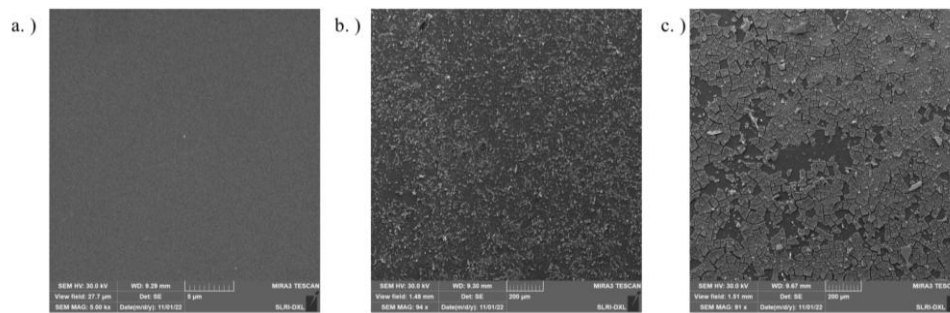


Figure 3. FE-SEM images of (a) FTO glass, (b) FTO/ TiO_2 and (c) FTO/ WO_3

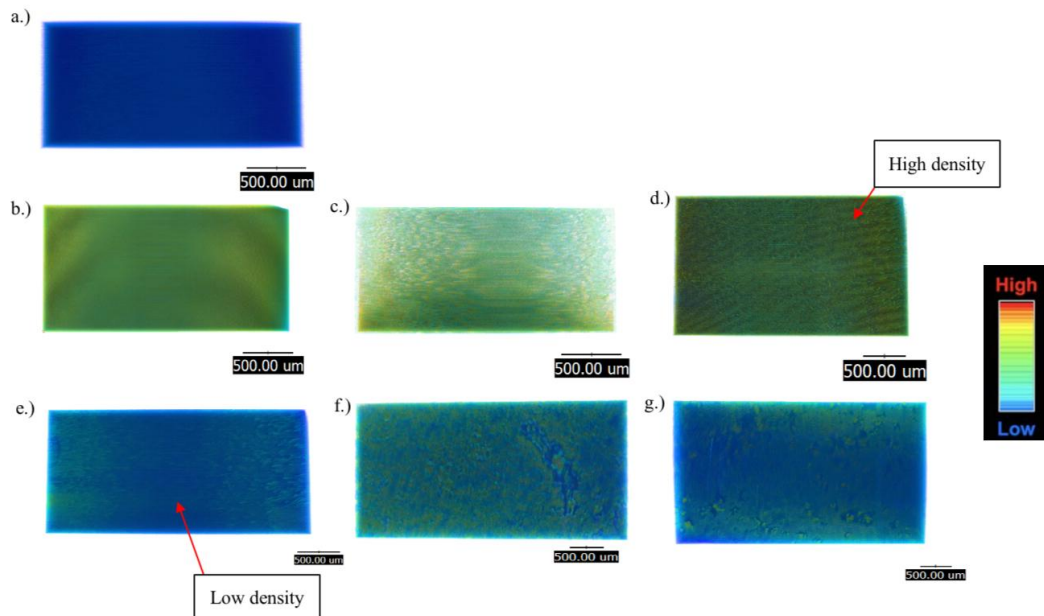


Figure 4. The 3D Reconstruction of 7 glasses with the gradient color for identify the density and the dipping particles (a) the FTO glass without dipping process, (b-d) the FTO with dipping TiO_2 particles for (b) 2 layers (c) 4 layers (d) 6 layers and (e-g) the FTO after dipping with WO_3 for (e) 2 layers (f) 4 layers (g) 6 layers, respectively

The uniformity and density of TiO₂ and WO₃ coated layer demonstrated by the SRXTM radiographic analysis technique as changes with the number of coating layers increases. This is a main factor that affects the ability of light absorbance and transmittance.

The visible light wavelength range between 380-750 nm was used to studied as the wavelength range in which both materials had high absorption and could be used in solar cell operating conditions. From Figure 5, It is found that the FTO conductive glass has low absorbance properties. After coating with TiO₂ and WO₃, the percentage of absorbance was higher with a tendency to increase directly proportional to the number of TiO₂ and WO₃ coated layer. From this result, FTO/WO₃ coated material has a higher absorbance than FTO/TiO₂ coated material. However, the distribution of the coated layer is uneven, resulting in low absorbance abnormalities for more layered coatings. While the light absorbance of FTO/TiO₂ coated material tends to significant rise as the number of layers increased and the absorbance of FTO/TiO₂ was steady

throughout with the condition of visible light analysis.

Figure 6 shows the light transmittance properties of FTO glass, FTO/TiO₂ and FTO/WO₃ coated material. The light transmittance property is opposite with light absorption property. Of course, the light transmittance of uncoated FTO conductive glass must be higher than that of FTO/TiO₂ and FTO/WO₃ coated material. Then, especially of FTO/TiO₂ and FTO/WO₃ coated material was considered on light transmittance property. From this result, FTO/TiO₂ coated material has a higher transmittance than FTO/WO₃ coated material. While the light absorbance of FTO/TiO₂ coated material tends to decrease significantly when the number of coated layers is increased. For FTO/WO₃ coated material, the light transmittance of FTO/WO₃ coated material is lower than FTO/TiO₂ coated material. It was also found that the abnormal transmittance property with insignificant increase in value when increase of coating number, due to the unevenness of coated layer.

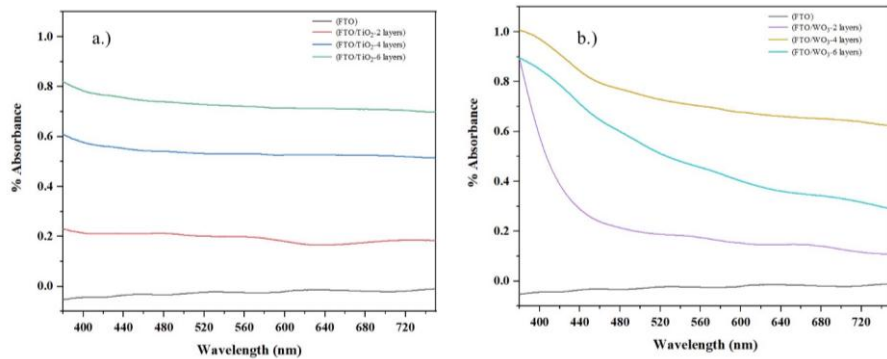


Figure 5. The light absorbance versus wavelength graph of FTO, (a) FTO/TiO₂ and (b) FTO/WO₃ at different number of coated layers

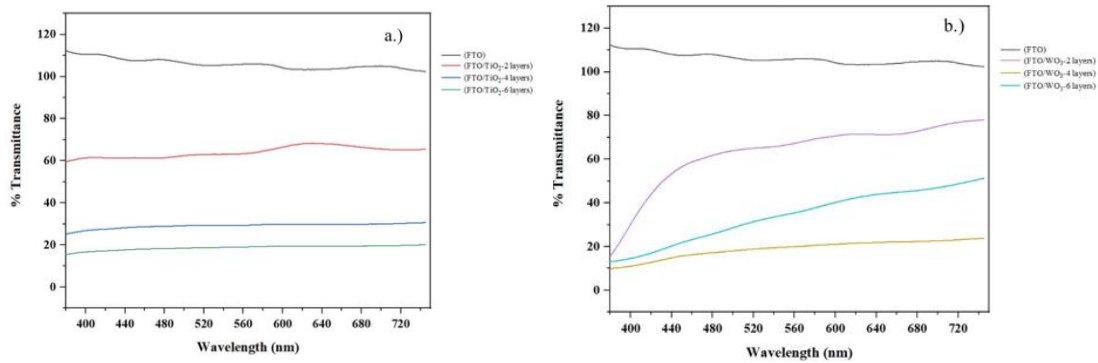


Figure 6. The light transmittance versus wavelength graph of FTO, (a) FTO/TiO₂ and (b) FTO/WO₃ at different number of coated layers

Conclusions

This work demonstrates that the SR-XTM technique can be used to analytical a uniformity and density of TiO₂ and WO₃ coated layers influencing absorbance and transmittance properties. The optical property test confirmed that the absorbance tends to increase with increasing of the coated layer density. The increasing of TiO₂ coated layer is more significantly increased the coated density. In addition, with the same experiment of WO₃ coated layer revealed that the uniformity and density of WO₃ coated layer were lower than those of TiO₂ coated layer. The unevenness of WO₃ coated layer caused the uneven absorbance, besides the uniformity of coated layer decrease as the number of coated layer increase. As the coated material is unevenly distributed, some areas are denser than others. From this result, The TiO₂ coated layer is most acceptable considered of uniformity coating layer that can be verified with the SR-XTM technique, even though the TiO₂ coated layer has a tendency to increase the absorbance less than WO₃ coated layer. Consequently, TiO₂ is more suitable for using as a coated material that need to increase their light absorption ability.

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