Synthesis and characterization of TiO$_2$ doped SnO$_2$ thin film prepared by sol-gel method

Devit Suriyani Che Halin$^{1*}$, Kamrosni Abdul Razak$^{1,2}$, Azliza Azani$^1$, Mohd Mustafa Al Bakri Abdullah$^1$, Mohd Arif Anuar Mohd Salleh$^1$, Norsuria Mahmed$^1$, Nur Syahirah Shafee$^1$, Muhammad Mahyiddin Ramli$^2$, Ayu Wazira Azhari$^3$, and Varishta Chobpattana$^4$

$^1$Center of Excellence Geopolymer & Green Technology (CEGeoGTech), School of Materials Engineering, Universiti Malaysia Perlis, (UniMAP), 02600 Jalan Kangar-Arau, Perlis.
$^2$School of Microelectronic Engineering, Universiti Malaysia Perlis, Pauh Putra Campus, 02600, Arau, Perlis, Malaysia.
$^3$Water Research Group (WAREG), School of Environmental Engineering, Universiti Malaysia Perlis 02600 Arau, Perlis, Malaysia.
$^4$Department of Materials and Metallurgical Engineering, Faculty of Engineering, Rajamangala University of Technology Thanyaburi (RMUTT), Thailand.

Abstract. In this work, preparation of titanium dioxide doped with tin oxide, SnO$_2$/TiO$_2$ thin films deposited onto silicon wafer via sol-gel method. Different amount of SnO$_2$ was added (5 ml, 10 ml and 15 ml) into parent solution. The obtained films were annealed at different temperature which is 400°C, 500°C and 600°C for 1 hour. Morphological and surface topography of the SnO$_2$ doped TiO$_2$ thin films were studied using Scanning Electron Microscope (SEM) and Atomic Force Microscope (AFM). The annealed films shows non-uniform crack due to the mismatch of coefficient of thermal expansion (CTE) between SnO$_2$/TiO$_2$ thin films and silicon wafer.

1 Introduction

Nowadays, there are numerous application of titanium dioxide (TiO$_2$) that benefits the society such as cosmetics, foods, restraining and sterilizing virus, electrical devices and photocatalytic oxidation (PCO) under ultraviolet (UV) and visible light also known as a pollution control technology [1, 2]. Self-cleaning activity will occur when TiO$_2$ is being exposed under light source which will display a photocatalytically induced superhydrophilicity properites. The hydrophobic surface of the substrate will transform to hydrophilic surface after coated with TiO$_2$ where it will cause a uniform water film. This will prevent the adhesion of organic or inorganic components on the surface. Therefore, the cleanliness can be retained. TiO$_2$ thin film can be coated on the surfaces of various substrates such as metals, textiles, ceramics, fibres and glass that will exhibits self-sterilisation and self-cleaning properties when it is exposed to the light source.

* Corresponding author : dewisuriyani@unimap.edu.my
When TiO$_2$ is doped with metal oxide such as SnO$_2$, it will improve the efficiency of TiO$_2$ photocatalyst. By doping SnO$_2$ into TiO$_2$ thin films, hydrophilicity and photocatalytic activity of composite thin films would be improved due to the reduction of TiO$_2$ particle growth rate [3-6].

In this work, TiO$_2$ doped with SnO$_2$ thin films coated on silicon substrate were fabricated using a sol-gel dip coating technique. The thin films were annealed at 400°C, 500°C and 600 °C for 1 hour with the heating rate of 5 °C/min. X-ray Diffractometer (XRD) was used to study the phase composition of SnO$_2$ doped TiO$_2$ thin films while Scanning Electron Microscope (SEM) and Atomic force microscope (AFM) was used to observe the morphology and the surface topography of SnO$_2$ doped TiO$_2$ thin films, respectively.

2 Methodology

2.1 Materials

The main raw materials used to produce TiO$_2$ sol in these studies are titanium (IV) isoproxide (TTIP), Ti(OC$_3$H$_7$)$_4$, isopropanol, C$_3$H$_8$O and acetic acid, CH$_3$COOH. Tin-tetrachloride pentahydrate (SnCl$_4$.5H$_2$O) powder was used as the source of SnO$_2$ solution. The silicon wafer was used as a substrate.

2.2 Methods

Firstly, 0.5 ml titanium (IV) isoproxide (TTIP) and 10 ml isopropanol, C$_3$H$_8$O were stirred at room temperature for 20 minutes using magnetic stirrer. Then, 2.5ml of acetic acid was added into the solution while stirring to speed up the chemical reaction. SnO$_2$ solution was produced by adding 50ml of distilled water and 50 ml of ethylene glycol into 3.51g tin-tetrachloride penta-hydrate (SnCl$_4$.5H$_2$O) powder. The solution was dissolved for 10 minutes to make a 100 ml precursor mixture. The TiO$_2$ solution was mixed together with 5ml, 10ml and 15ml of SnO$_2$ respectively. The mixture was vigorously stirred for another 30 minutes at room temperature. The substrate used is silicon wafer (1 cm x 1 cm). The silicon wafers were washed in acetone using ultrasonic cleaner for 30 minutes. The TiO$_2$/SnO$_2$ sol were deposited on silicon wafer by using spin coating at 1500rpm for 30s. The coated samples were dried at 40°C for 10 minutes. After that, the coated samples were annealed in muffle furnace for 1 hour with heating rate of 5°C/min at 400°C, 500°C and 600°C. The TiO$_2$ doped with SnO$_2$ thin films were prepared by sol-gel method according to the amount of SnO$_2$ doping. In this study, the characterization of TiO$_2$ thin films was investigated by using Scanning Electron Microscope (SEM) and Atomic Force Microscopes (AFM).

3 Results and Discussions

Fig. 1 shows the XRD pattern of SnO$_2$ doped TiO$_2$ thin films. It can be seen that the diffraction pattern shows dual-phase of anatase and rutile with no existence of SnO$_2$. The samples were annealed at three different temperatures which are, 400°C, 500°C and 600°C, respectively. The angle 2θ shifts towards lower angles with the increase of annealing temperature from 400 °C to 600 °C.
Fig. 1. XRD pattern of SnO$_2$ doped TiO$_2$ thin films which was annealed at 400°C, 500°C and 600°C.

Fig. 2 and Fig. 3 show the micrographs of SnO$_2$/TiO$_2$ thin films that were coated on silicon wafer with different amount of SnO$_2$ added and annealed in muffle furnace at 400°C, 500°C and 600°C for 1 hour. The micrographs show a non-uniform films with large flaky and large cracks film. It might due to the surface tension between the film and the air during the drying process [7]. It can be seen that as the annealing temperature increases, the distance between cracks become smaller. The higher the annealing temperature, the least cracks formed. To prevent crack from forming when annealed at high temperature, more layers of coating were deposited to ensure the coverage of substrate become better and formed a thicker film. It is well agree with the previous work by Abdul Razak et al [8]. From Fig. 2, it was observed that 5 ml SnO$_2$ doped with TiO$_2$ annealed at 400°C has the most agglomerated region as compared to the samples annealed at 500°C and 600°C. Meanwhile, in Fig. 3 shows the micrograph of 10 ml SnO$_2$ doped with TiO$_2$ annealed at 500°C has the most agglomerated region as compared to the samples annealed at 400°C and 600°C. The micrographs show a Volmer-Weber growth. Volmer–Weber growth of SnO$_2$ doped TiO$_2$ thin films includes nucleation of 3-D islands, growth, impingement and coalescence of islands [4].

The surface topography of SnO$_2$/TiO$_2$ thin films was investigated by Atomic Force Microscopes (AFM). Typical topographical images of the surface of SnO$_2$/TiO$_2$ coatings are represented in 3D images. Fig. 4 shows the surface topography of 5ml SnO$_2$ doped TiO$_2$ thin films at different annealing temperature of 400°C, 500°C and 600°C respectively. Fig. 5 shows the surface topography of 10ml SnO$_2$ doped TiO$_2$ thin films at different annealing temperature 400°C, 500°C and 600°C respectively.
Fig. 2. SEM image of the surface morphology for 5 ml SnO$_2$ doped with TiO$_2$ thin films at temperature of (a) 400°C, (b) 500°C and (c) 600°C.

RMS=1.155nm

RMS=3.677nm

RMS=3.796nm

Fig. 3. SEM image of the surface morphology for 10 ml SnO$_2$ doped with TiO$_2$ thin film at temperature of (a) 400°C, (b) 500°C and (c) 600°C.

Fig. 4. The surface topography and 3D images of 5ml SnO$_2$ dope TiO$_2$ thin films at different annealing temperature of (a) 400°C, (b) 500°C and (c) 600°C.

RMS=1.570nm

RMS=2.995nm

RMS=9.937nm

Fig. 5. The surface topography and 3D images of 10 ml SnO$_2$ dope TiO$_2$ thin films at different annealing temperature of (a) 400°C, (b) 500°C and (c) 600°C.

It can be observed that the sample which was annealed at 600°C produce the highest value of roughness (RMS) while the sample annealed at 400°C produced the lowest value of roughness (RMS). This showed that the surface roughness (RMS) of the samples
increased with the annealing temperature. It is well agreed that the presences of SnO₂ influenced the RMS value. As the amount of SnO₂ added increased, the RMS value also increased. This revealed that the RMS value was affected by annealing temperature and the amount of SnO₂ added into the parent solution.

4 Conclusion

In this study, SnO₂ doped TiO₂ thin films were prepared by sol-gel method. The XRD pattern shows the phase composition of SnO₂ doped TiO₂ thin films were dual-phase of anatase and rutile with no existence of SnO₂. The morphological characteristics of SnO₂ doped TiO₂ thin films have non-uniform cracks, agglomerate and the formation of Volmer-Weber growth. When the annealing temperature and the amount of SnO₂ increased, the surface roughness (RMS) value increased.

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