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Synthesis and Characterization of TiO₂/SiO₂ Thin Film via Sol-Gel Method

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Abstract. TiO₂/SiO₂ thin films were prepared by sol-gel spin coating method. Structural, surface morphology and optical properties were investigated for different annealing temperatures at 300°C, 400°C and 500°C. X-ray diffraction pattern show that brookite TiO₂ crystalline phase with SiO₂ phase presence at 300°C. At higher temperatures of 400-500°C, the only phase presence was brookite. The surface morphology of film was characterized by scanning electron microscopy (SEM). The films annealed at 300°C shows an agglomeration of small flaky with crack free. When the temperature of annealing increase to 400-500°C, the films with large flaky and large cracks film were formed which was due to surface tension between the film and the air during the drying process. The UV-Vis spectroscopy shows that the film exhibits a low transmittance around 30% which was due to the substrate is inhomogeneously covered by the films. In order to improve the coverage of the film on the substrate, it has to repeatable the spin coating to ensure the substrate is fully covered by the films.

1. Introduction

TiO₂ is a material which have its chemical stability and electronic properties and well known as semiconductor.

It has been used in wide application in semiconductor electrochemistry, solar energy conversion, as gas sensor, in electronic devices, and as antireflective coatings [1]. TiO₂ becomes one of the most popular photocatalytic materials.

The TiO₂ naturally have existing in several crystalline phase such as rutile, brookite, and anatase. Among of these, anatase and rutile crystalline phase are widely used in application of solar cell. Rutile phase are suitable for protective coating on lense due to stable thermodynamically and high refractive



index. At lower temperatures, brookite and anatase phases are more stable however both will change to the rutile phase at certain temperature. Due to the photocatalytic activity, most of the researches tend to use anatase, TiO_2 . The larger band gap in crystalline phase in anatase because it has better response to ultraviolet photons for photocatalysis.

Photocatalytic is the process in which the solar energy is converted into electrical energy in the solar cell. The photocatalytic performance of these compounds depends on the characteristics of the TiO_2 crystallites, such as the size and surface area [2].

Therefore, modification of its physical and chemical property is of interest to researchers [3,4]. One of the possible ways to modify the property of TiO_2 crystallites is by adding a second semiconductor into the TiO_2 matrix. Silicon dioxide (SiO_2) has been incorporated into the TiO_2 matrix to enhance the photocatalytic process [5-8].

SiO_2 has high thermal stability, excellent mechanical strength and helps to create new catalytic active sites due to interaction between TiO_2 and SiO_2 [9].

Recently, Zhou et al. [8] demonstrated that mixed metal oxides (TiO_2 - SiO_2) enhance the photocatalytic performance due to improved surface adsorption and increasing surface hydroxyl group in the thin film. Also, at the same time SiO_2 acts as the carrier of TiO_2 and helps to obtain a large surface area as well as a suitable porous structure [10].

It has been proven by the researchers that by adding SiO_2 as a adding material to the TiO_2 can improve the photocatalytic activity and hydrophilicity process [11-13].

Based on previous researchers stated, additions of SiO_2 to the TiO_2 can prevent the formation of rutile phase at high temperature up to 800°C that is the undesired phase for the photocatalytic applications in the solar cell [14].

This research work is focus on synthesized and characterized the structural, morphology and optical properties of $\text{TiO}_2/\text{SiO}_2$ thin film with three different annealing temperatures in order to improve the properties the self-cleaning property in solar applications.

2. Methodology

The sol-gel method was used to synthesize $\text{TiO}_2/\text{SiO}_2$ thin films. The tetraethyl orthosilicate, TEOS, and tetra-n-butyl orthotitanate, TBOT, were used as the sources of SiO_2 and TiO_2 respectively to prepare the solutions of SiO_2 solutions and TiO_2 solutions.

Firstly, TBOT was mixed together with ethanol and deionized water by using molar ratio of mixture (TBOT: ethanol: deionized water) was 1:0.5:3. Next, hydrochloric acid was added drop by drop into the mixing solutions which give the function as a catalyst. Then, this solution was mixing together for 1 hour by using magnetic stirrer.

After 1 hour, a clear solution was obtained then TEOS was added into the solution until the ration of TBOT: TEOS was 1:1. Then, the solution was continued stirred for 30 minutes at 45°C . The final solutions was undergoes aging process for 24 hours. Figure 1 shows the flowchart of the synthesized $\text{TiO}_2/\text{SiO}_2$ thin film.

Thin films of $\text{TiO}_2/\text{SiO}_2$ were deposited by the method of spin coating in air at room temperature, on glass substrate with speed at 1000 rpm for 30s. Finally, the films were annealed at 300°C , 400°C and 500°C in air for 3 hours by using muffle furnace.

The crystalline structure was characterized by an X-ray diffractometer (XRD) in 2θ range from 20° to 80° . Scanning electron microscopy (SEM) was used to study the surface morphology and pore distribution of the produced films.

The transmission and absorbance were measure by UV/VIS Perkin-Elmer spectrophotometer.

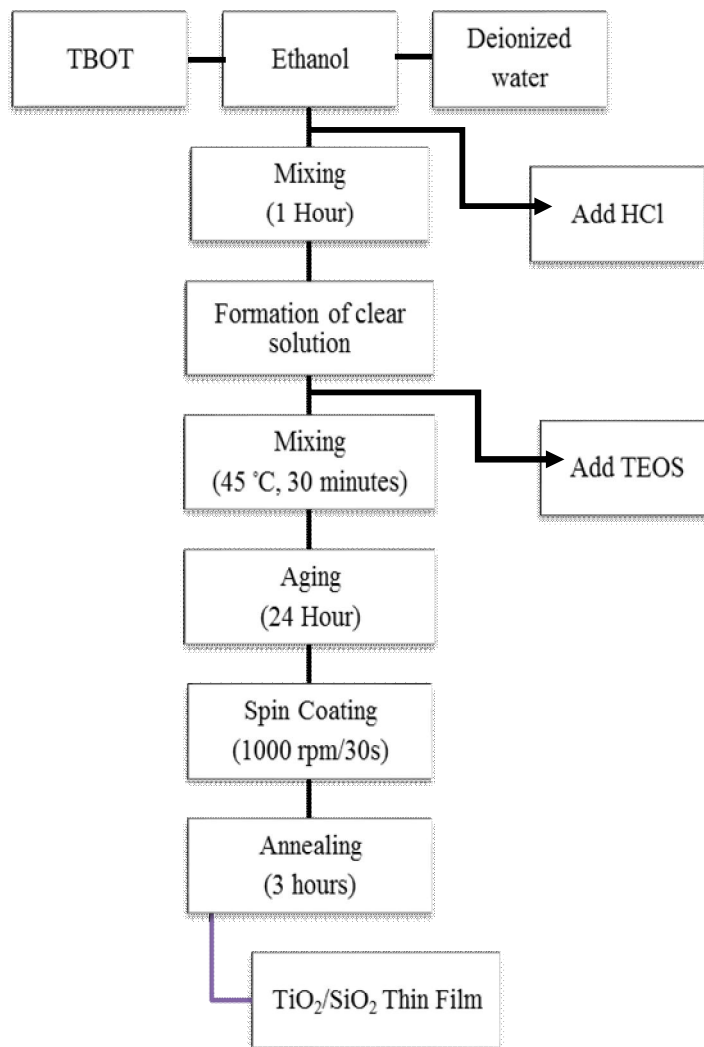


Figure 1. The flowchart of the synthesized TiO₂/SiO₂ composite thin film.

3. Results and discussion

X-ray diffraction pattern has been used to investigate the phase of the prepared TiO₂/SiO₂ films. The X-ray diffraction pattern of TiO₂/SiO₂ thin films annealed at different temperatures is shown in figure 2. The peaks have been indexed and found to be that of brookite TiO₂ and SiO₂. The diffraction peak at $2\theta = 31.99^\circ$ was assigned to the characteristic diffraction peak of brookite TiO₂ phase and corresponds to (1 2 1) direction. For the SiO₂ phase, the diffraction peak at $2\theta = 66.54^\circ$ which is corresponds to (3 0 0) direction. The diffraction pattern shows that an annealing at a temperature equal or lower than 300°C would be largely sufficient to form TiO₂/SiO₂ completely. It is well agreed with Maeda & Yamasaki [15] which found that when the films was annealed at 250°C for 180 min, the phase of titania–silica (TiO₂-SiO₂) mixed films exist. They also reported on addition of SiO₂ into the TiO₂ films can prevents the recrystallization of the TiO₂ composites. When the temperature of annealing was increase to 400°C and 500°C, the only peak reveal was brookite TiO₂ films. Mechiakh et al. [16] was reported, at higher

temperatures (400°C and 450°C), in addition to anatase TiO₂ the formation of brookite TiO₂ which crystallizes with the (1 2 1) plane parallel to the surface. No peak corresponding to the SiO₂ phase is seen in the diffraction pattern of films annealed at 400°C and 500°C.

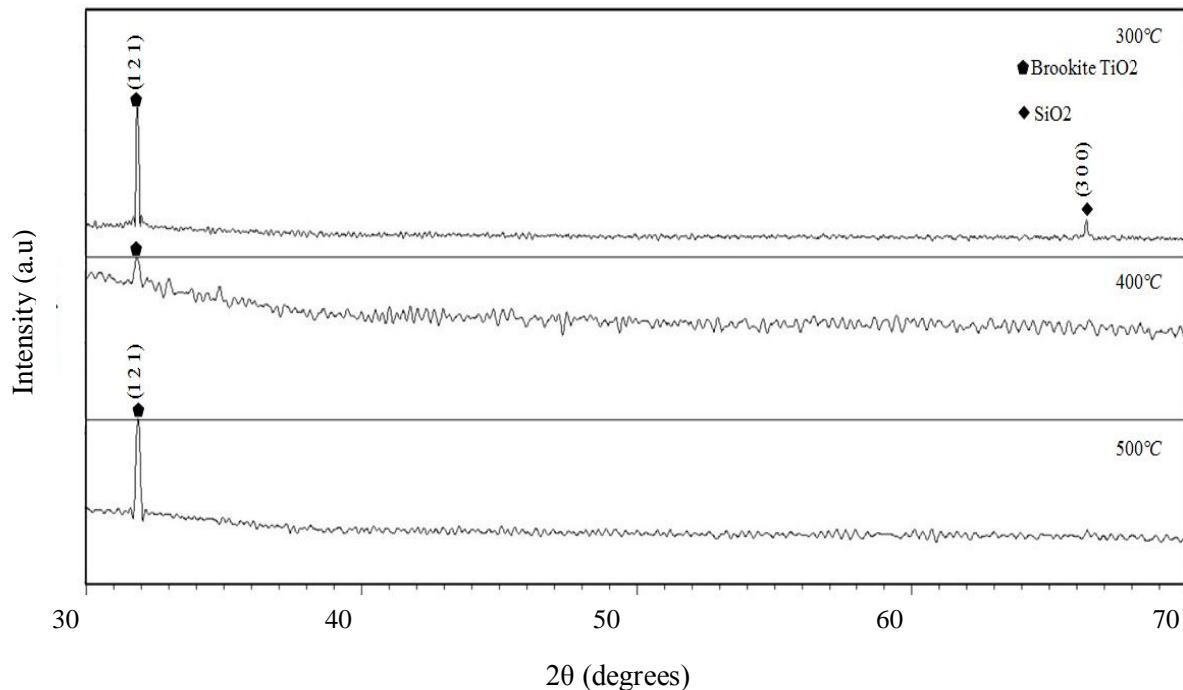


Figure 2. X-ray diffraction pattern of TiO₂/SiO₂ films annealed at different temperatures.

Many efforts have been made to modify the TiO₂ by impurities for improving the photocatalytic activity under visible light [17-19]. Modification of TiO₂ with SiO₂ has been attracted in the past decades and widely used due to photocatalysis and hydrophilicity properties [20]. The characteristic of SiO₂ itself, such as its water-trapping effect, is suggested to play an important role for surface hydrophilicity of the TiO₂/SiO₂ films. In the TiO₂/SiO₂ films, therefore, the synergistic effects of anatase polycrystalline structure of the TiO₂ and water-trapping effect of the SiO₂ are important for their strong photo-induced hydrophilicity [15].

Figure 3 shows the SEM micrographs of TiO₂/SiO₂ annealed at 300°C, 400°C and 500°C. The SEM micrographs revealed that the surface morphology of the TiO₂/SiO₂ films depend strongly upon annealing temperature. It was observed that the average grain size of the films increases significantly when annealing temperature increases. In the presence of SiO₂ for the films annealed at 300°C (figure 3(a)), it can be seen many nucleation centers are present on the substrate and agglomeration of small flaky are produced. Then, the films with small flaky size are not able to grow into bigger ones and prevent crack. Whereas for higher annealing temperatures as seen in figure 3(b) and (c) it revealed large flaky size and large cracks throughout the coatings. Temperature increasing may lead to non-uniform and cracked coating and reduce coating lucidity and transparency [21].

It also clearly seen that the substrate is inhomogeneously covered by large flaky size and it may formed during drying process due to surface tension between the film and the air [22]. In the case of sol-gel coated films, during drying process, the capillary forces might have generated which provides cracks on the surface [23].

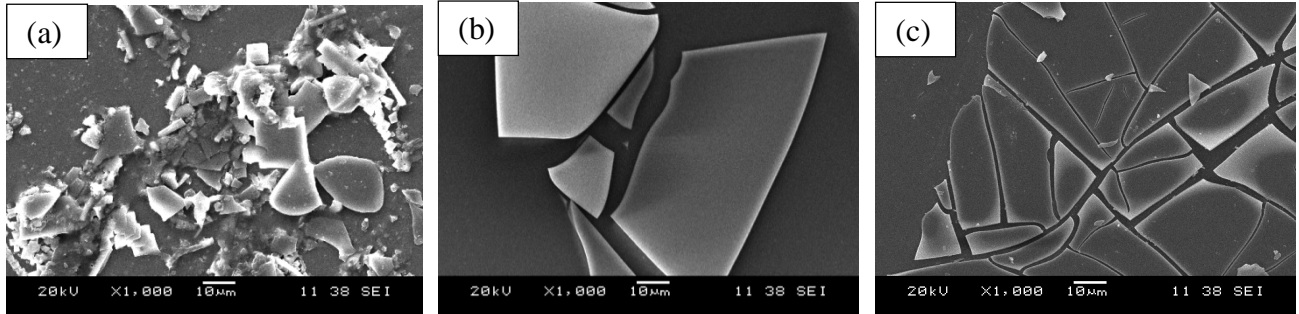
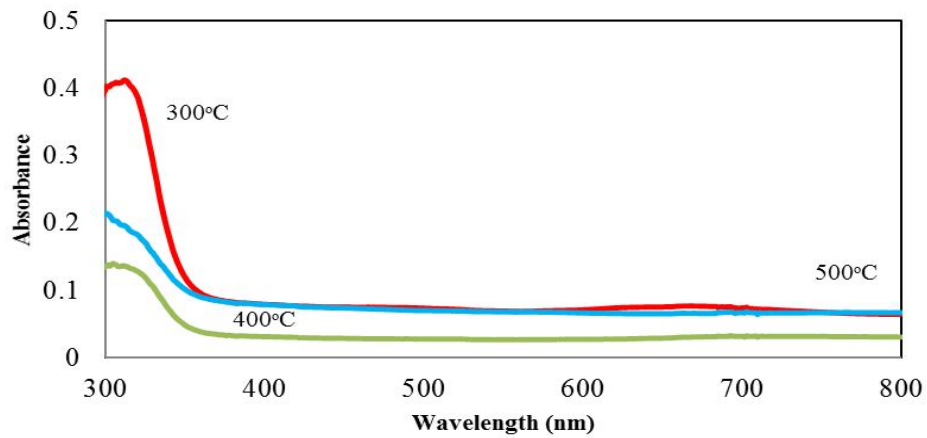
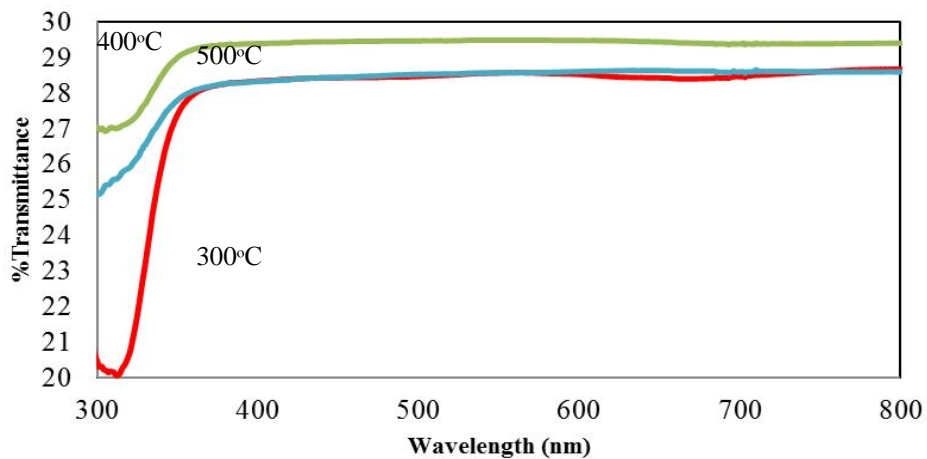


Figure 3. SEM micrographs of TiO₂/SiO₂ annealed at (a) 300°C, (b) 400°C and (c) 500°C.



(a)



(b)

Figure 4. UV-Vis spectra of absorbance and transmittance for TiO₂/SiO₂ films annealed at different temperatures.

The optical absorbance and transmittance spectra of TiO₂/SiO₂ films deposited on glass substrates with different annealing temperatures were shown in figure 4(a) and (b). The films show sharp absorption edge in ultraviolet region at a wavelength of about 320 nm as seen in figure 4(a). The absorbance is much higher for the films which annealed at 300°C with the formation of TiO₂/SiO₂ and the films are crack free. The transmittance within the visible and near infrared region is lower than 30% including the glass substrates, which reveals the poor optical properties of TiO₂/SiO₂ produced in this study. It is well agreement with the SEM micrograph which the substrate was not fully covered with the films and the films have large cracks. When the films are having large cracks, the UV light will be scattered in all direction.

Optical transparency is prerequisite for the application of self-cleaning coating on transparent glass or plastic surface. The films annealed at 400°C and 500°C shows the same trends of spectra for absorbance and transmittance with the only brookite phase exist. The transmittance of film annealed at 400°C is much higher compared to the film annealed at 500°C is due to pure TiO₂ film at 400°C is having less crack and bigger crystal size. The increased of cracking crystal size is responsible for the slight loss in optical transmission in the visible range. The process of spin coating needs to repeat a few times, in order to ensure the substrate is fully covered by the films and it also can enhance the transmittance and the absorbance of the films [24].

4. Conclusions

This work deals with the preparation and characterization of TiO₂/SiO₂ thin films using a simple and cost effective method: sol-gel spin coating. The deposition was performed on glass substrate at room temperature and annealed at three different annealing temperatures; 300°C, 400°C and 500°C. This work presents the annealing temperature effect on the properties of TiO₂/SiO₂. X-ray diffraction pattern shows that the thin films obtained at annealing temperature 300°C, having a phase of TiO₂/SiO₂. At higher annealing temperature (400-500°C), the formation of brookite phase presence. The SEM micrograph of the film annealed at 300°C show that the film consist of agglomeration of small flaky size without any crack. The analysis of UV-Vis transmission spectra shows that TiO₂/SiO₂ thin films are transparent in the visible range with a poor transmittance due to large crack and non-uniform film. This research proved that annealing temperature is an important parameter which affects the structure; surface morphology, and optical properties of the films.

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