

## Research article

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### TiO<sub>2</sub>/SiO<sub>2</sub> Coated 310S Stainless Steel for Hydrogen Peroxide Generation via Photocatalytic Reaction

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Received: 2 February 2021, Revised: 18 June 2021, Accepted: 11 August 2021

DOI: 10.55003/cast.2022.03.22.001

#### Abstract

##### Keywords

TiO<sub>2</sub>/SiO<sub>2</sub>;  
photocatalysis;  
hydrogen peroxide

The photocatalytic reactor was designed by assembling the parts of a photocatalyst, an ultraviolet light source and an air distribution. This prototype reactor was used to generate H<sub>2</sub>O<sub>2</sub> via photocatalytic reaction. TiO<sub>2</sub> and TiO<sub>2</sub>/SiO<sub>2</sub> prepared from sol-gel method were coated on 310S stainless steel substrate using dip-coating technique. Rice husk ash was used as the source of SiO<sub>2</sub>. The major phase of all the pure TiO<sub>2</sub> and SiO<sub>2</sub> particles was anatase and amorphous, respectively. The surface analysis showed the coated TiO<sub>2</sub> and TiO<sub>2</sub>/SiO<sub>2</sub> with an average roughness of 365.391 and 1322.001 nm, respectively, compared with the average thickness of coated TiO<sub>2</sub> which was measured to be 2.7325 μm and for TiO<sub>2</sub>/SiO<sub>2</sub>, which was 4.7856 μm. The results also showed the coating of TiO<sub>2</sub>/SiO<sub>2</sub> was highly porous at the surface compared with pure TiO<sub>2</sub>. The TiO<sub>2</sub>/SiO<sub>2</sub> show good photocatalytic reaction activity because SiO<sub>2</sub> doping on TiO<sub>2</sub> increased the adsorption of O<sub>2</sub> and H<sub>2</sub>O molecules towards the photoactive center of TiO<sub>2</sub>. Thus, the incorporation of SiO<sub>2</sub> onto the photocatalyst helped to increase photocatalytic activity. The generation of H<sub>2</sub>O<sub>2</sub> increased with increasing ambient humidity up to 60% RH, but then decreased. This behavior was explained by the effect of water vapor adsorption on the SiO<sub>2</sub> surface. The air flowrate enhanced the generation of H<sub>2</sub>O<sub>2</sub>, higher air flow rate produced a higher amount of H<sub>2</sub>O<sub>2</sub>. H<sub>2</sub>O<sub>2</sub> was generated at the highest concentration of 64 ppmv from the TiO<sub>2</sub>/SiO<sub>2</sub> photocatalyst at ambient humidity of 60% RH and air flowrate 8.20 m/s.

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## 1. Introduction

The pandemic of COVID-19 is now the most challenging issue of mankind across the world. It was reported by World Health Organization (WHO), as of 8 June 2021, that the number of infections had reached 173 million confirmed cases [1]. The disinfection of pathogenic viruses, fungi, bacteria, molds, endospores, biofilms, organic and inorganic contaminants is now an interesting research area [2]. On the surfaces and in the atmosphere, these pathogens from various sources are widely distributed. There are various ways for disinfection such as physical and chemical methods [3]. Due to the mentioned problems, it is very interesting to design new sterilization processes that can kill or eliminate these microorganisms via an air purifier. However, the feasibility of such technology and processes must be carefully studied to ensure efficiency, safety and sustainability. Photocatalytic oxidation process is one way to destroy microorganisms and involves oxidizing organic contaminants [4].

One of the main members of the reactive oxygen species (ROS) group of molecules is hydrogen peroxide ( $H_2O_2$ ), which is commonly considered to have antimicrobial properties, and tends to be an abundant molecule. Hydrogen peroxide is normally present in food and can be found in drinking water, rainwater and sea water [5-8]. Moreover, our lungs have enzymes that continuously convert oxygen and moisture in the lungs to hydrogen peroxide [9]. Hydrogen peroxide has been used for disinfection and microbial control in aqueous solutions. In today's efforts to use hydrogen peroxide gas, because the distribution of molecules in the gas phase is better than in liquid phase and it is particularly useful when the low concentration is available for operating. Recently, there has been a lot of effort to use hydrogen peroxide gas as the gas molecule is more able to diffuse through systems even at low concentration. Due to its rapid effectiveness, low temperature operation, surface compatibility and minimal toxicity issues, hydrogen peroxide gas-based processes have become common alternatives to other chemical and physical antimicrobial methods [10]. In addition, the Occupational Safety and Health Administration (OSHA) recommends that the safety level of vaporized hydrogen peroxide concentration lower than 1.0 ppm [11].

Most of the research concerning photocatalysis have been focused on oxidation processes, and the generation of hydrogen peroxide for use as an oxidizing agent [12]. Numerous research studies of photocatalytic processes have reported on the generation of hydrogen peroxide produced from hydrogen and oxygen from mixtures of water and alcohol, using ultraviolet light and a photocatalyst [13, 14]. In recent years, a wide range of photocatalyst, including  $TiO_2$ ,  $ZnO$ ,  $SnO_2$ ,  $WO_3$ ,  $CdS$ ,  $ZrO_2$ ,  $Sb_2O_4$  and  $Fe_2O_3$  have been investigated for their photocatalytic efficiency [15]. Titanium dioxide, in particular, have received a lot of research interest due to their catalytic activity. In order to increase the photocatalytic activity of  $TiO_2$ , the integration of  $TiO_2$  with other agents, especially  $SiO_2$ , was found to induce reactant molecules to stay on the surface, a factor that when combined with sufficient ultraviolet light led to the reaction occurring more readily [16].

Several works have been studied on the use of  $TiO_2/SiO_2$  photocatalyst in photocatalytic processes, such as the photocatalytic degradation of organophosphorus pesticides [17-26]. To synthesize the photocatalyst, the sol-gel technique was reported to be a simple and cost-effective process and this solution can be further used for coatings [27-30]. An important factor that influences photocatalytic reaction efficiency is the photocatalytic reactor's design because photocatalytic efficiency is highly dependent on light absorption. The immobilization of the catalyst onto a substrate is the basis of photocatalytic reactors for air reactions. Surface hydroxyl content, film thickness, and substrates all impact the gas-solid reaction that occurs on the catalyst thin film [31-33]. The flat plate reactor is the most often utilized gas phase reactor [34-36]. Thus, 310S stainless steel plates are of great interest. They can serve as catalyst support materials in photocatalytic reactors for air reactions. The stainless steel 310S is made from austenitic heat resistant alloy with

excellent oxidation resistance at 1093.33°C under mildly cyclic conditions and allow better air flow than quartz plates and other materials.

In this work, the dip-coating of 310S stainless steel substrates with  $\text{TiO}_2/\text{SiO}_2$  prepared by the sol-gel method were developed to use as photocatalyst. The prepared photocatalyst was used as part of the assembly of a prototype device to generate hydrogen peroxide gas from ambient humidity air via photocatalytic reaction. Rice husk ash was used as the source of silicon dioxide. The physical and chemical characteristics of  $\text{TiO}_2$  and  $\text{TiO}_2/\text{SiO}_2$  catalysts on 310S stainless steel were investigated. The concentration of generated hydrogen peroxide was measured with variation of air relative humidity and air flowrate.

## **2. Materials and Methods**

### **2.1 Preparation of $\text{TiO}_2$ and $\text{TiO}_2/\text{SiO}_2$ photocatalyst**

#### **2.1.1 Chemicals**

Titanium (IV) butoxide reagent grade (97% (v/v) Sigma), ethanol absolute grade (99.9% (v/v)), sulfuric acid (98% (v/v)) and hydrochloric acid (37% (v/v) of ANaPURE) were purchased from New Zealand. Nitric acid (65% (v/v)), acetylacetone and hydrogen peroxide (30% (v/v)) were purchased from CARLO ERBA reagents, France. Rice husks were obtained from the local rice mill located in Nakhon Ratchasima, Thailand.

#### **2.1.2 Preparation of amorphous $\text{SiO}_2$ powder**

Amorphous  $\text{SiO}_2$  powder was prepared as previously published [16]. First, a bulk 100g of rice husk, rich in silica, was rinsed in 1 l of sulfuric acid solution with concentration of 0.5 M at 80°C for 2 h, then rinsed again with water until pH equal to 7 was obtained. After rinsing, the rice husk was dried at 105°C for 24 h, then calcined in furnace with the heating rate set at 5°C/min, holding at 600°C, for 2 h. The obtained product was amorphous  $\text{SiO}_2$ .

#### **2.1.3 Preparation of $\text{TiO}_2$ and $\text{TiO}_2/\text{SiO}_2$ photocatalyst solution via sol-gel method**

$\text{TiO}_2$  and  $\text{TiO}_2/\text{SiO}_2$  catalysts were prepared via the Sol-gel method. The preparation of  $\text{TiO}_2$  photocatalyst solution was started by mixing the titanium (IV) n-butoxide 68 ml with ethanol absolute 210 ml,  $\text{HNO}_3$  2.56 ml, DI water 5.5 ml and acetylacetone 10.4 ml. For the preparation of  $\text{TiO}_2/\text{SiO}_2$  photocatalyst solution, the fixed ratio of 1:0.76% (w/w) of  $\text{TiO}_2:\text{SiO}_2$  was used [16].  $\text{SiO}_2$  prepared from rice husk was added. Then the solution was stirred for 8 h at 200 rpm and at 80°C. After stirring, the solution was further left for 24 h to achieve the gel solution and it was a liquid and solid solution mixed.

### **2.2 Preparation of photocatalyst coated 310S stainless steel**

#### **2.2.1 Coating of the 310S stainless steel substrates**

Each 310S stainless steel plate had a diameter of 130 mm with a thickness of 1 mm. Each plate was perforated with hole sizes of 1 mm across the plane surface. The first step was the surface treatment of the stainless steel to ensure maximum adhesion between the coating layer and the substrate.

During this process, all plates were immersed in molar 1:1 with sulfuric acid and hydrogen peroxide for 30 min. Each plate was then rinsed with DI water. Finally, the plates were dried at 100°C.

The dip coating technique was selected for use in this work. Each plate was initially covered by tape on the smooth surface near the edge of the plate. Thus, after coating, the photocatalyst was attached on the surface zone of perforation. After coating, the plate was left to dry overnight at room temperature. Finally, the dried coated plates were calcined at temperature of 600°C for 2 h.

The sols were filled with powders prepared by firing gels of the same composition, with the maximum thickness for preparing thicker layers. Until a homogeneous mixture was obtained, these powders were introduced into the sols under stirring. This technique allows good quality thick layers to be prepared because the shrinkage during the drying stage was significantly reduced.

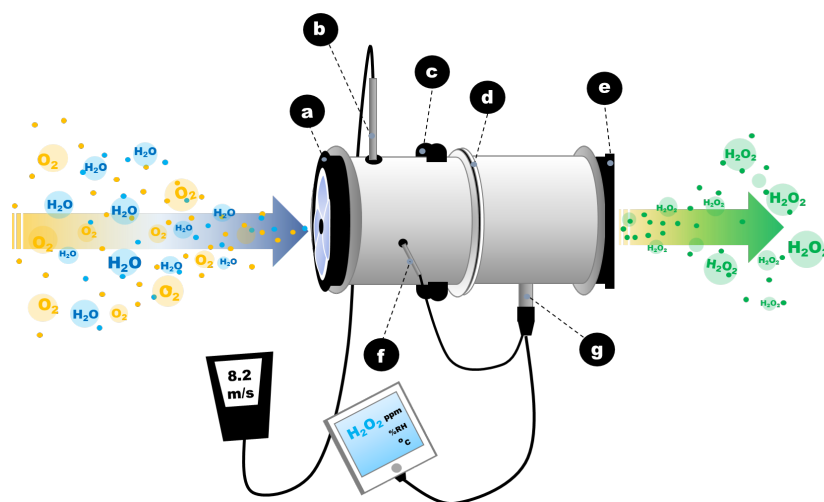
### 2.2.2 Characterization of catalysts

X-ray diffraction measurements (XRD) were carried out using a traditional Bragg-Brentano diffractometer with Ni-filtered Cu K $\alpha$  radiation to identify the crystalline phases of the heat-treated powder samples. A field emission scanning electron microscope (FESEM) and energy dispersive X-ray spectroscopy (EDS) were used to analyze the materials' morphologies. Measurements of roughness and thickness of TiO<sub>2</sub> and TiO<sub>2</sub>/SiO<sub>2</sub> films were performed using a Bruker Contour GT profiler, which was used to measure thickness at each point in the field of view, a process that illustrated thickness variations.

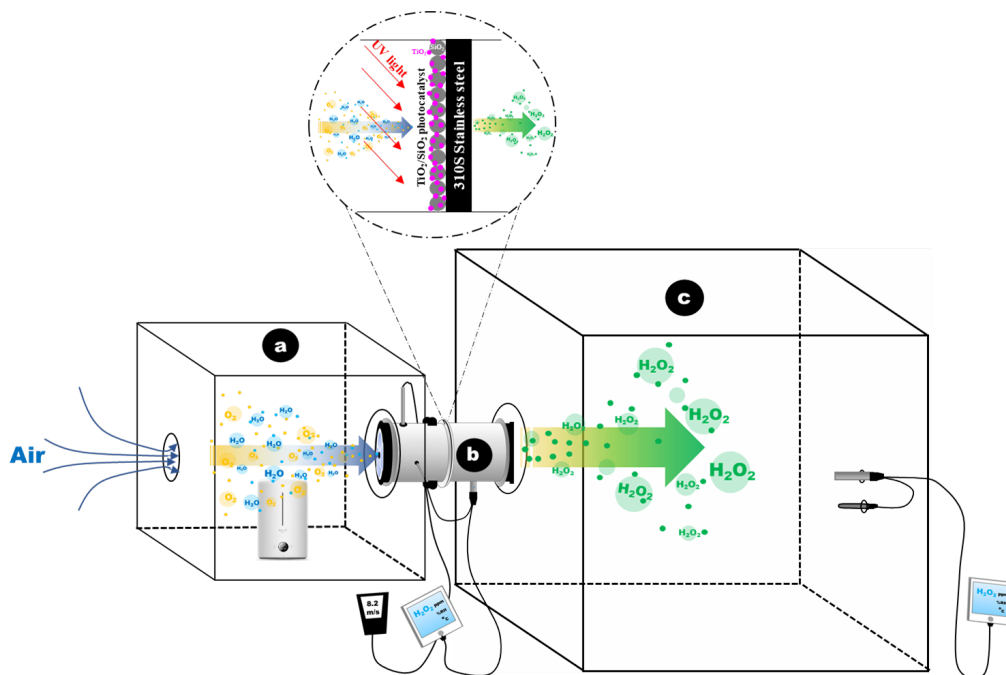
## 2.3 Hydrogen peroxide generator

### 2.3.1 Photocatalytic reactor system and its operating method

A prototype of the photocatalytic reactor was assembled, as shown in Figure 1. The frame is an acrylic cylindrical vessel with length of 240 mm, inside diameter of 100 mm, and the thickness of 10 mm. The stainless steel plate which was coated with photocatalyst, was placed vertically inside the horizontal vessel. The UV lamp was inserted inside the vessel and fixed near the front of photocatalyst plate. A small fan with adjustable flow rate of air was attached in the front of the vessel. A device for measurement of the air flow rate was placed inside the vessel and put in after the fan. A probe to measure the generated hydrogen peroxide was inserted in the vessel and it was put after the photocatalyst plate. This probe was a model HPP272 supplied by Vaisala indigo 210, Finland. A smaller fan was also put at the end of the vessel to blow out the generated product gas to the surroundings. For the relative humidity variation, a control box, as shown in Figure 2 (a), was connected with the reactor vessel. The air humidifier was put inside this box and used for controlling the relative humidity. In Figure 2 (c), this box was connected with the reactor to simulate a surrounding volume to measure the distribution of generated hydrogen peroxide.



**Figure 1.** Schematic of photocatalytic reactor for hydrogen peroxide generation  
(a) fan, (b) air flow meter, (c) UV light source, (d) 310S stainless steel plate, (e) ventilation fan, (g) hydrogen peroxide meter, and (f) ambient temperature humidity meter



**Figure 2.** Schematic of (a) chamber for controlling humidity of air, (b) prototype photocatalytic reactor, and (c) chamber for measuring hydrogen peroxide distribution

### 3. Results and Discussion

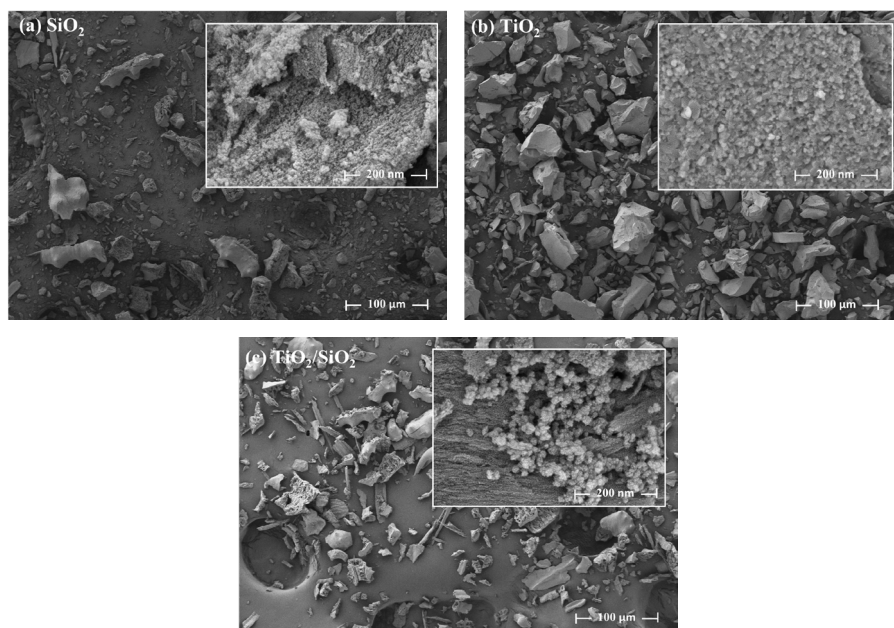
#### 3.1 Characterization of $\text{TiO}_2$ and $\text{TiO}_2/\text{SiO}_2$ photocatalyst

##### 3.1.1 Analysis of photocatalysts synthesis

Figure 3 shows the FESEM micrographs of  $\text{SiO}_2$ ,  $\text{TiO}_2$  and  $\text{TiO}_2/\text{SiO}_2$  powder. Apparently,  $\text{SiO}_2$  had a rougher surface structure than that of  $\text{TiO}_2$ . The porous structure of  $\text{SiO}_2$  was created by removing the existing organic molecules as volatile chemicals [37]. The resulting structure of  $\text{SiO}_2$  after calcination at  $600^\circ\text{C}$  for 2 h created porous  $\text{SiO}_2$  composite (Figure 3a). The  $\text{TiO}_2$  crystal surface was quite smooth (Figure 3b). The synthesis of  $\text{TiO}_2/\text{SiO}_2$  photocatalysts with the dispersion of  $\text{SiO}_2$  on the surface increased the porosity of the structure (Figure 3c).

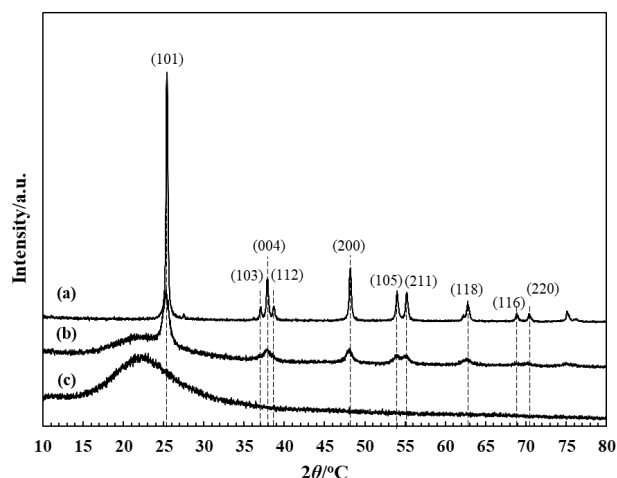
In Figure 4, after calcination at  $600^\circ\text{C}$  for 2 h, XRD patterns and crystalline phase content (anatase type  $\text{TiO}_2$ ) were observed. Anatase (JCPDS card no. 84-1286) peaks were found at  $2\theta$  values of  $25.4^\circ$ ,  $37.0^\circ$ ,  $38.0^\circ$ ,  $38.8^\circ$ ,  $48.0^\circ$ ,  $54.0^\circ$ ,  $55.2^\circ$ ,  $62.9^\circ$ ,  $68.9^\circ$  and  $70.5^\circ$ . These corresponded to the (101), (103), (004), (112), (200), (105), (211), (118), (116), and (220) crystal planes. No diffraction peak of silica was observed in the XRD patterns, showing that silica in rice husk ash was still amorphous.

Figure 5 shows the FESEM/EDS images of  $\text{SiO}_2$ ,  $\text{TiO}_2$  and  $\text{TiO}_2/\text{SiO}_2$  powder. The analyzed elements are O, Si, and Ti. In Figure 5a, the  $\text{SiO}_2$  analysis showed the average weight and atomic percentage of Si and O were 42.69 and 57.31 wt.%, 29.81 and 70.19 ac.%, respectively. Figure 5b, the  $\text{TiO}_2$  analysis indicated the average weight and atomic percentage of Ti and O were 75.77 and 24.23 wt.%, 51.10 and 48.90 ac.%, respectively. In Figure 5c, the  $\text{TiO}_2/\text{SiO}_2$  analysis showed the average weight and atomic percentage of Ti, Si and O were 41.63, 19.98 and 38.39 wt.%, 21.87, 17.87 and 60.26 ac.%, respectively. These results indicated that  $\text{TiO}_2$  particles were well dispersed with the  $\text{SiO}_2$ .

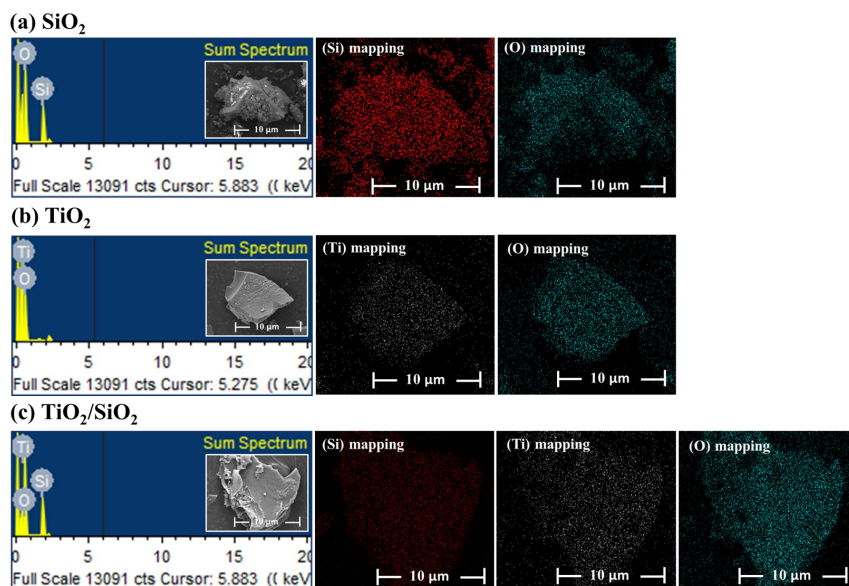


**Figure 3.** FESEM micrograph of samples (a)  $\text{SiO}_2$ , (b)  $\text{TiO}_2$ , and (c)  $\text{TiO}_2/\text{SiO}_2$  powder





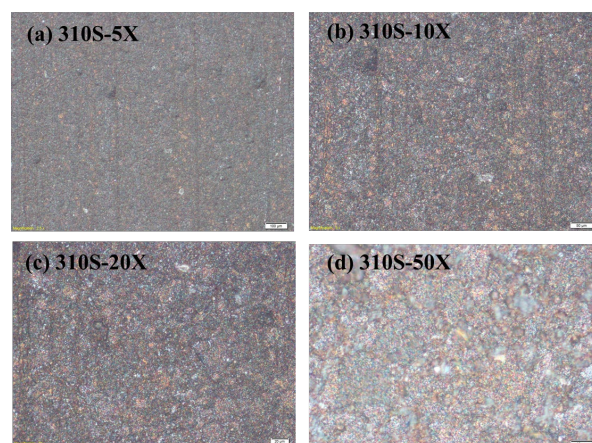
**Figure 4.** XRD patterns of photocatalysts (a)  $\text{TiO}_2$ , (b)  $\text{TiO}_2/\text{SiO}_2$ , and (c)  $\text{SiO}_2$



**Figure 5.** FESEM/EDS images of samples (a)  $\text{SiO}_2$ , (b)  $\text{TiO}_2$ , and (c)  $\text{TiO}_2/\text{SiO}_2$  powder

### 3.1.2 Surface analysis and 301S stainless steel coated photocatalysts

The chemical composition of the uncoated 301S stainless steel used in this study (in wt.%) was provided by the supplier and consisted of Cr 24.8, Ni 19.1, C 0.069, Si 0.75, Mn 1.37, P 0.036, S <0.001, O 0.013, Cu 0.07, Al 0.07, Ti 0.010, N 0.062 and the balance Fe [38]. The uncoated surface was characterized and it was found that the surface was quite smooth, as show in Figure 6.



**Figure 6.** Microstructure of the uncoated 310S stainless steel

Studies on the surface roughness of uncoated and coated  $\text{TiO}_2$  and  $\text{TiO}_2/\text{SiO}_2$  photocatalyst on 310S stainless steel surface were done. During measurements, two-dimensional and three-dimensional parameters of surface roughness were also determined, and height ( $Sa$ ,  $Sk_u$ ,  $Sp$ ,  $Sq$ ,  $Ssk$ ,  $Sv$ ,  $Sz$ ) was included. In fact, the topographic roughness of  $\text{TiO}_2/\text{SiO}_2$  was the greatest, a finding reported in Table 1 that confirmed our qualitative findings. It was computed for both average roughness ( $Sa$ ) ( $\sim 1322$  nm) and the root mean square roughness ( $Sq$ ) ( $\sim 1556$  nm). Furthermore, the topographic pattern is comparable for maximum peak height ( $Sp$ ), maximum pit height ( $Sv$ ), and maximum height ( $Sz$ ), indicating that topographic patterns in  $\text{TiO}_2/\text{SiO}_2$  are significantly different from  $\text{TiO}_2$  and uncoated photocatalysts. The asymmetry, or dissimilarity to a Gaussian distribution, is determined by the surface skewness ( $Ssk$ ), which is related to the third momentum of the height distribution. Positive  $Ssk$ , such as turned surfaces, have relatively high spikes that protrude above a flatter average. Negatively  $Ssk$ , a feature of porous surfaces, contains relatively deep valleys inside a smoother plateau [39]. The surface kurtosis ( $Sk_u$ ) value is a measurement of the roughness profile sharpness, above the mean plane, and the height distribution is skewed ( $Sk_u < 3$ ). The distribution of heights was normal (Parts that are sharp and parts that are indented coexist) ( $Sk_u = 3$ ), and height distribution was spiked ( $Sk_u > 3$ ). Figure 7 shows the surface roughness in 2D and 3D. The average roughness of the uncoated surface was 310.981 nm. The surface roughness of coated  $\text{TiO}_2$  and  $\text{TiO}_2/\text{SiO}_2$  were approximately 365.391 nm and 1322.001 nm, respectively. From these results, it indicates that coating with  $\text{TiO}_2/\text{SiO}_2$  gave the surface higher thickness and surface roughness as well as porosity, as previously mentioned.

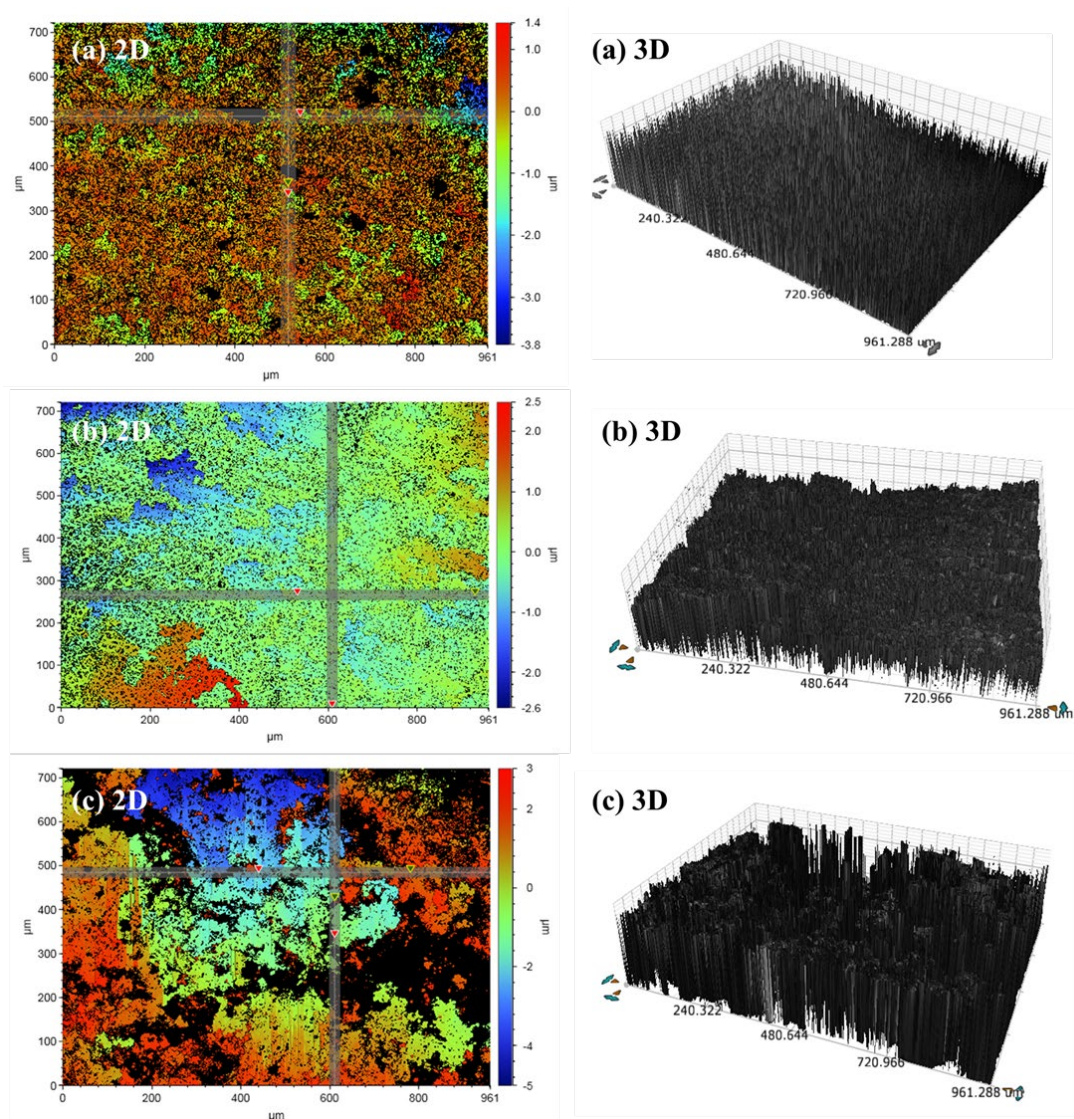
Figure 8 shows 2D and 3D analyses of the thickness of coated surface with different photocatalysts. The coated thickness of  $\text{TiO}_2$  and  $\text{TiO}_2/\text{SiO}_2$  were found to be 2.7325  $\mu\text{m}$  and 4.7856  $\mu\text{m}$ , respectively.

The microstructural characteristics of the coated 310S stainless steel with photocatalyst via the sol-gel using dip-coating technique were analyzed. The SEM images of the coated surface for  $\text{TiO}_2$  and  $\text{TiO}_2/\text{SiO}_2$  photocatalyst coating with different magnitudes are shown in Figure 9. As seen from Figure 9 (a)-(d), the  $\text{TiO}_2$  coated surface possesses a regular crystal morphology and quite smooth surface. The average size of crystal is about 16.13  $\mu\text{m}$ . The  $\text{TiO}_2/\text{SiO}_2$  coated surface can be seen in Figure 9 (e)-(h), and this surface exhibits a porous surface structure. It was found that the average size of crystal was about 14.47  $\mu\text{m}$  and the porous size was approximately 372.88 nm. However, due to the burning-out of organic compounds from rice husk, these vaporized molecules left the porous structure. Therefore, a network of pores was created in the silica surface.

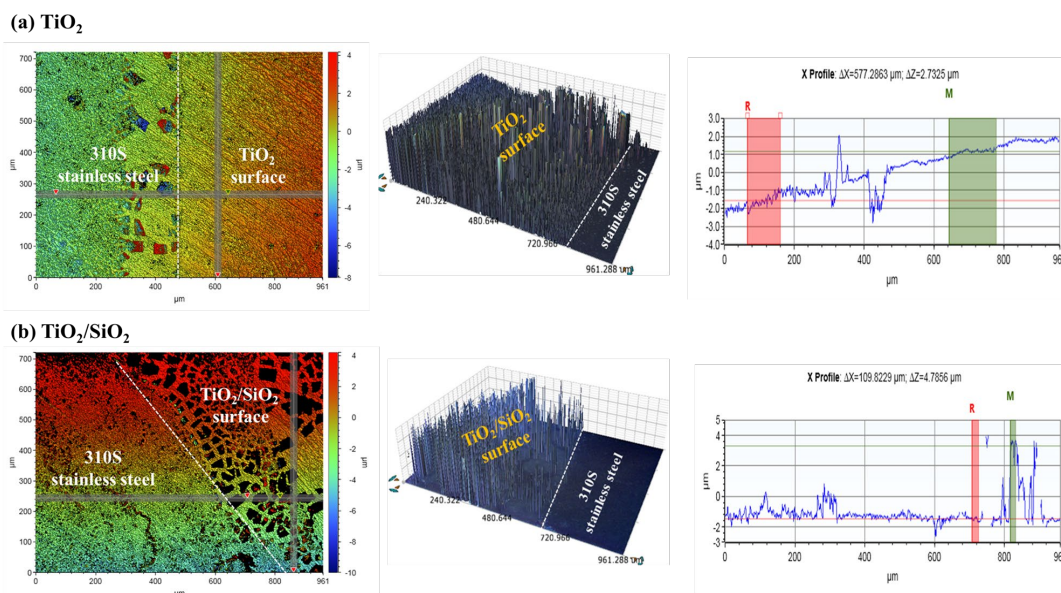


**Table 1.** Value of the 2D and 3D surface roughness parameters

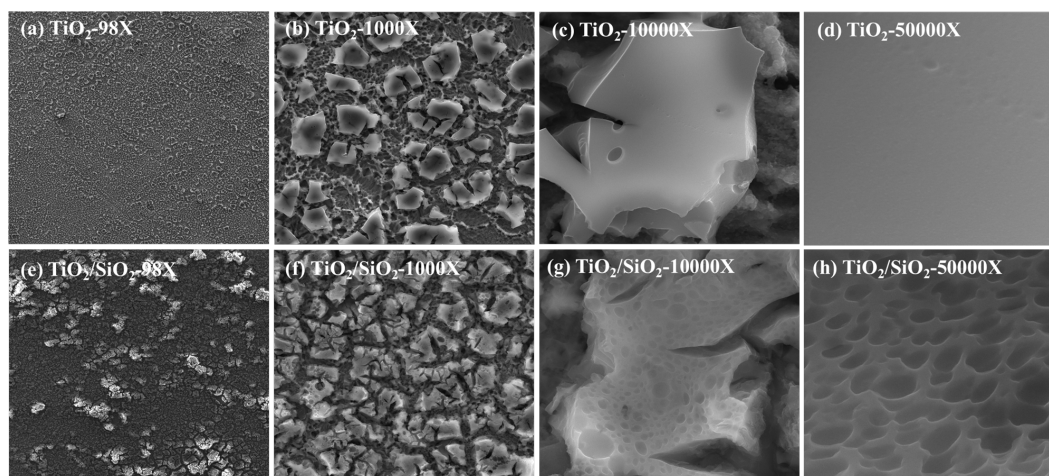
310S Sample	2D and 3D						
	<i>Sa</i> (nm)	<i>Sku</i> –	<i>Sp</i> (nm)	<i>Sq</i> (nm)	<i>Ssk</i> –	<i>Sv</i> (nm)	<i>Sz</i> (nm)
Uncoated	310.981	10.426	1378.222	439.913	-1.978	-3415.035	4793.257
Coated TiO <sub>2</sub>	365.391	4.915	2230.068	501.739	0.520	-2293.282	4523.350
Coated TiO <sub>2</sub> /SiO <sub>2</sub>	1322.001	2.093	3938.382	1556.645	0.192	-3479.549	7417.930



**Figure 7.** Surface roughness with 2D and 3D  
(a) uncoated photocatalysts (b) coated TiO<sub>2</sub> (c) coated TiO<sub>2</sub>/SiO<sub>2</sub>



**Figure 8.** Show the thickness after coating  $\text{TiO}_2$  and  $\text{TiO}_2/\text{SiO}_2$  photocatalysts

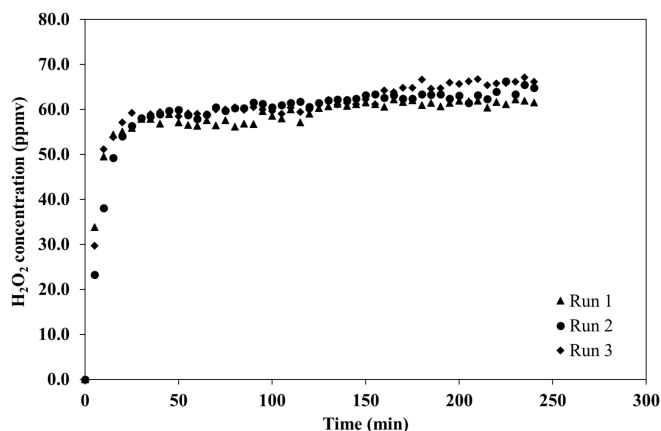


**Figure 9.** SEM micrograph of a  $\text{TiO}_2$  and  $\text{TiO}_2/\text{SiO}_2$  photocatalyst coated on 310S stainless steel

## 3.2 Photocatalytic reactor for hydrogen peroxide gas generation

### 3.2.1 Dry hydrogen peroxide generation with $\text{TiO}_2$ and $\text{TiO}_2/\text{SiO}_2$ photocatalyst

Figure 10 shows the time courses photocatalytic reactor operation and concentration of dry hydrogen peroxide generation with UV light, ambient humidity of air 60% RH, temperature 30°C and  $\text{TiO}_2/\text{SiO}_2$  photocatalyst for photocatalytic reactor tests the same condition. Photocatalytic reactor testing for hydrogen peroxide synthesis was done to ensure that the reactor can synthesize in the same conditions. The test was performed three times.



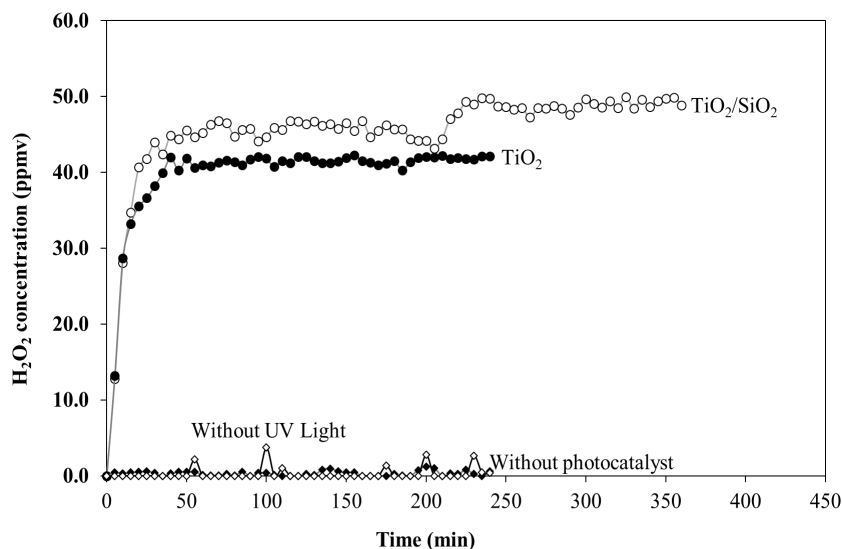
**Figure 10.** Time courses photocatalytic reactor operation and concentration of dry hydrogen peroxide generation with UV light, ambient humidity of air 60% RH, temperature 30°C, air velocity of 8.2 m/s, and TiO<sub>2</sub>/SiO<sub>2</sub> photocatalyst for photocatalytic reactor tests

Figure 11 shows the time courses for the photocatalytic reactor operation and concentration of dry hydrogen peroxide generation with UV light, air flow rate 5.1 m/s, ambient humidity of air 60% RH and temperature 30°C of a comparison of TiO<sub>2</sub> and TiO<sub>2</sub>/SiO<sub>2</sub> coated surfaces. The blank tests were run for without UV light and without photocatalyst, and as expected, no reaction occurred. However, a low concentration of dry hydrogen peroxide was detected due to its existence in the environment [5-9]. The TiO<sub>2</sub>/SiO<sub>2</sub> photocatalyst appeared to give higher concentrations of hydrogen peroxide than the TiO<sub>2</sub> photocatalyst. Thus, the incorporation of SiO<sub>2</sub> into the photocatalyst helped increase photocatalytic activity. Due to the simultaneous functions of TiO<sub>2</sub> and SiO<sub>2</sub> as photocatalyst and adsorbent binary mixed oxide, TiO<sub>2</sub>/SiO<sub>2</sub> proved to be a superior photocatalyst to TiO<sub>2</sub> alone. According to the mechanism shown in Figure 12, the addition of SiO<sub>2</sub> to the structure of TiO<sub>2</sub> significantly improved the specific surface area of TiO<sub>2</sub> based photocatalysts. Because of its high porosity, TiO<sub>2</sub>/SiO<sub>2</sub> had a large surface area. Structures with high porosity have a large internal surface area per unit mass, which gives great accessibility and diffusivity, allowing molecules to pass through pores, resulting in increase in the adsorbed molecules breakdown on the catalyst's surface. The doping of SiO<sub>2</sub> on TiO<sub>2</sub> increased the adsorption of O<sub>2</sub> and H<sub>2</sub>O molecules towards the photoactive center of TiO<sub>2</sub>, and more molecules were broken down, resulting in a rapid degradation rate under UV light.

### 3.2.2 Relationship between ambient humidity and dry hydrogen peroxide generation with TiO<sub>2</sub>/SiO<sub>2</sub> photocatalyst

Figure 13 shows the time course photocatalytic reactor operation and concentration of dry hydrogen peroxide generation with UV light, air flow rate 5.1 m/s, temperature 30°C and TiO<sub>2</sub>/SiO<sub>2</sub> photocatalyst for different relative humidity. The humidity fed into photocatalytic reactor has an effect on the photocatalytic reaction for hydrogen peroxide generation. It was found that increasing the relative humidity up to 60% increased the concentration of generated hydrogen peroxide. However, at the 70% RH the concentration of generated hydrogen peroxide was lower than that at 60% RH. The results may have been due to the effect of water adsorption by the SiO<sub>2</sub> porous structure as represented in Figure 14. In Figure 14 (a)-(b), at the low relative humidity, molecules of water were mostly trapped or adsorbed inside the pore of SiO<sub>2</sub>. Thus, there was a small portion





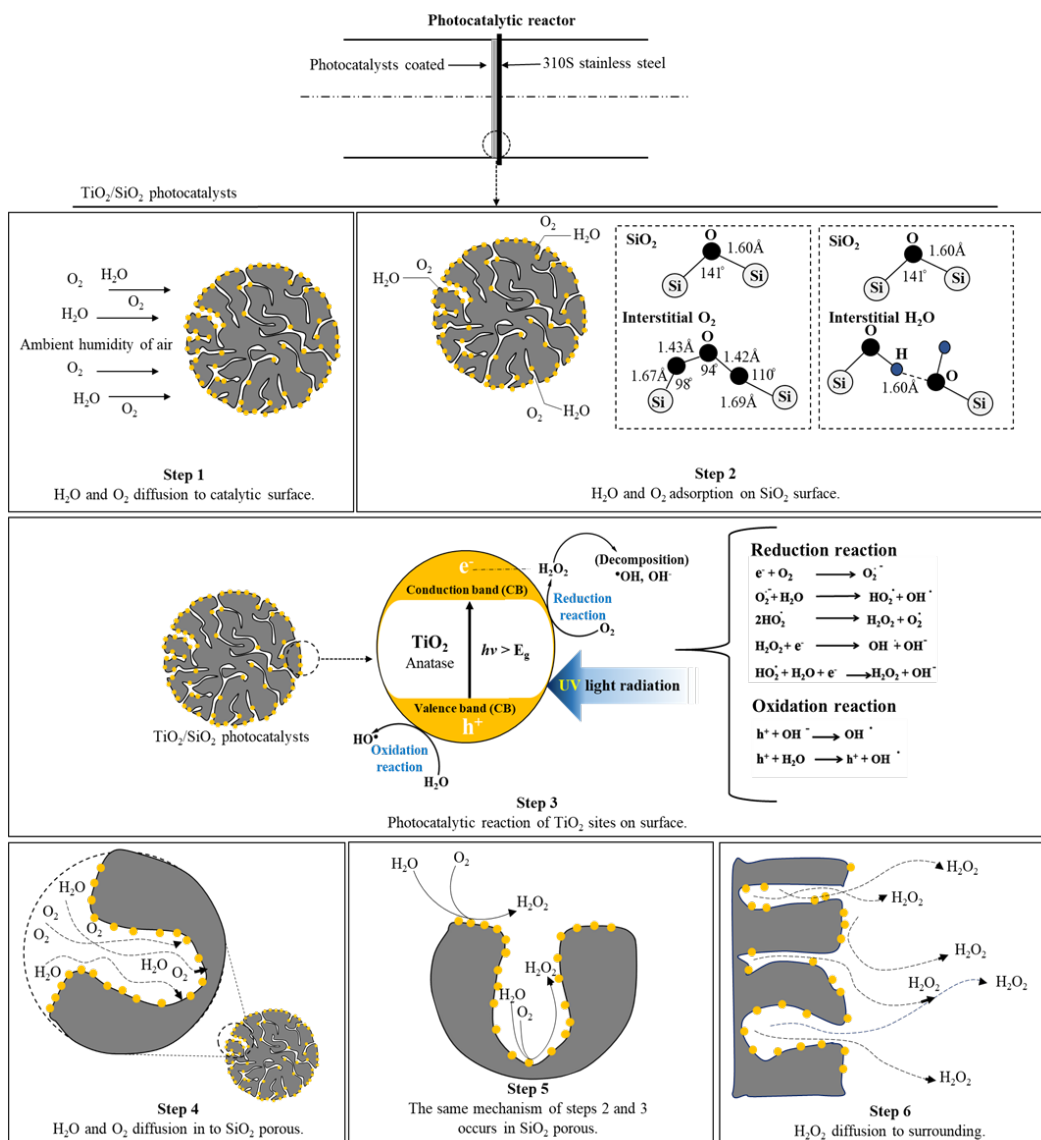
**Figure 11.** Time courses photocatalytic reactor operation and concentration of dry hydrogen peroxide generation with UV light, air flow rate 5.1 m/s, ambient humidity of air 60% RH, and temperature 30°C for TiO<sub>2</sub> and TiO<sub>2</sub>/SiO<sub>2</sub>

of the free water molecules left in the bulk stream to react by photocatalytic mechanism and become hydrogen peroxide. When the relative humidity reached 60% (Figure 14 (c)), the SiO<sub>2</sub>'s pores were full with water molecules. The portion of free water molecules in bulk phase was quite large, and these molecules were reacted to generate hydrogen peroxide. However, when the relative humidity was at 70% (Figure 14 (d)), the portion of free water molecules in bulk phase was excessive, leading to some water molecules covering or accumulating on the active sites. These active sites then became inactive, and the concentration of generated hydrogen peroxide decreased.

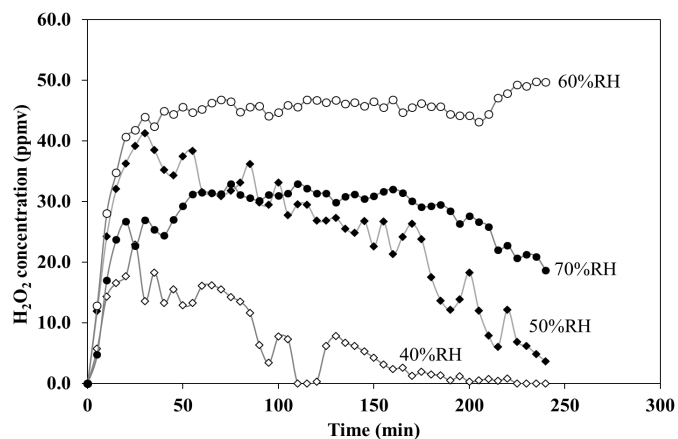
### 3.2.3 Relationship between velocity of ambient humidity and dry hydrogen peroxide generation with TiO<sub>2</sub>/SiO<sub>2</sub> photocatalyst

The time course for photocatalytic reactor operation and concentration of dry hydrogen peroxide generation for different air flowrate with UV light, ambient humidity of air 60% RH, temperature 30°C and TiO<sub>2</sub>/SiO<sub>2</sub> photocatalyst is shown in Figure 15. It was found that increasing in the air flowrate increased the concentration of generated hydrogen peroxide. This trend may have been due to the higher flowrate giving a higher feed amount of water molecules into the system. Thus, a higher number of water molecules was photocatalyzed to hydrogen peroxide. The high flowrate also reduced film resistance at the active sites. From this reason, all active sites demonstrated good photocatalytic activity.

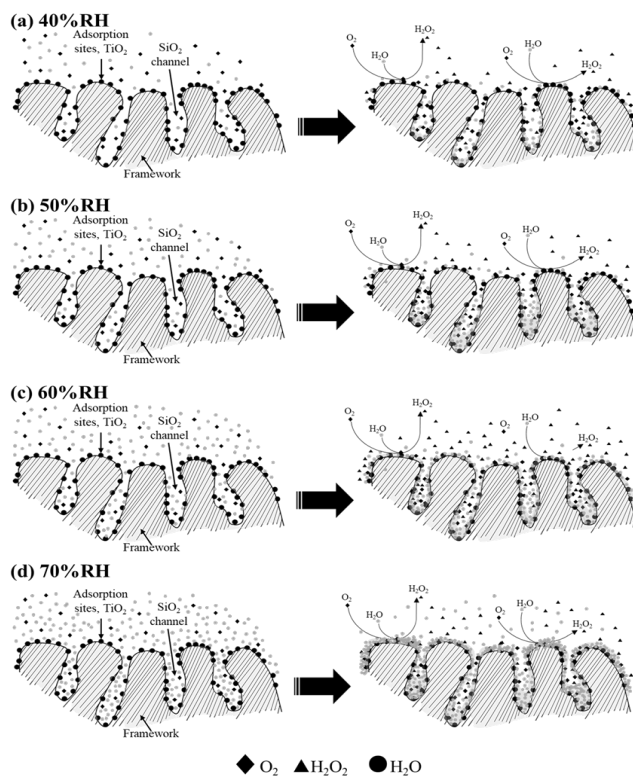
A comparison of this research with other studies regarding the photocatalytic production of hydrogen peroxide with TiO<sub>2</sub> based photocatalysts is shown in Table 2. In this study, hydrogen peroxide in gaseous form was synthesized at concentrations up to 64 ppmv (1.88 mM) after 1 h.



**Figure 12.** Mechanisms of photocatalytic reaction for hydrogen peroxide generation

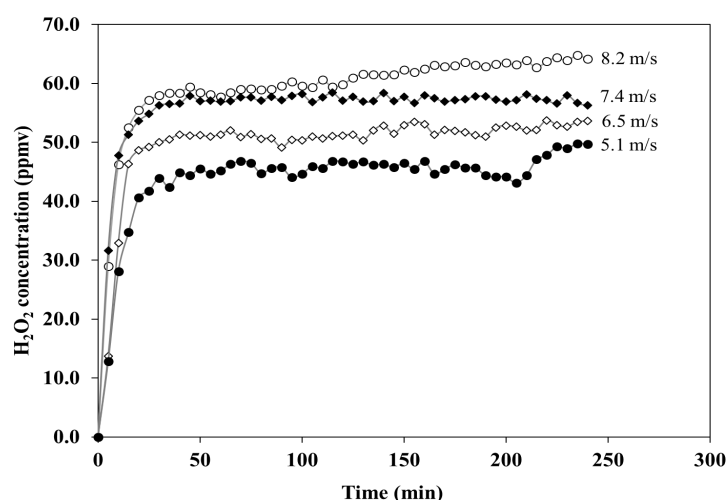


**Figure 13.** Time course photocatalytic reactor operation and concentration of dry hydrogen peroxide generation with UV light, air flow rate 5.1 m/s, temperature 30°C and TiO<sub>2</sub>/SiO<sub>2</sub> photocatalyst for different relative humidities



**Figure 14.** Schematic representation of the water adsorption in SiO<sub>2</sub>'s porosity and portion of free water molecules in bulk phase





**Figure 15.** Time courses photocatalytic reactor operation and concentration of dry hydrogen peroxide generation with UV light, ambient humidity of air 60% RH, temperature 30°C and TiO<sub>2</sub>/SiO<sub>2</sub> photocatalyst

**Table 2.** Summary of the photocatalytic production of H<sub>2</sub>O<sub>2</sub> with TiO<sub>2</sub> based photocatalysts

Material	Sacrificial reagent	Irradiation conditions	H <sub>2</sub> O <sub>2</sub> yield (1h)	Reference
TiO <sub>2</sub> /SiO <sub>2</sub> (in this work)	Ambient humidity	UV	1.88 mM	-
Au/TiO <sub>2</sub>	HCOOH	$\lambda > 420$ nm	0.64-0.70 mM	[40]
TiO <sub>2</sub> /rGO/carbon dots	2-propanol	AM 1.5	ca. 0.35 mM	[41]
TiO <sub>2</sub> /WO <sub>3</sub> /rGO	2-propanol	AM 1.5	ca. 0.27 mM	[42]
SN-GQD/TiO <sub>2</sub>	2-propanol	$\lambda > 420$ nm	0.451 mM	[43]
Co@TiO <sub>2</sub>	CH <sub>3</sub> OH	$\lambda = 400$ nm	1.7 mM	[44]
SNG/TiO <sub>2</sub>	2-propanol	$\lambda \geq 300$ nm	0.373 mM	[45]
Pt/TiO <sub>2</sub>	-	Full spectrum	5.096 mM	[46]

#### 4. Conclusions

TiO<sub>2</sub> and TiO<sub>2</sub>/SiO<sub>2</sub> coatings were prepared by the sol-gel method and dip-coating technique on 310S stainless steel substrates. It was verified by SEM that the coating was highly porous on surface TiO<sub>2</sub>/SiO<sub>2</sub> photocatalyst. The prototype photocatalytic reactor was designed by assembling the parts of photocatalyst, ultraviolet light source and air distribution system. The TiO<sub>2</sub>/SiO<sub>2</sub> photocatalyst irradiated with UV light generated a high concentration of H<sub>2</sub>O<sub>2</sub> from water vapor. The generation of dry hydrogen peroxide increased with increasing ambient humidity up to 60% RH, but then a decreasing trend was observed. This behavior was explained by the effect of water vapor adsorption on the SiO<sub>2</sub> surface. Air flowrate enhanced the generation of dry hydrogen peroxide, and higher air flow rate produced higher amounts of dry hydrogen peroxide. From all the tests in this investigation,

the prototype of photocatalytic reactor was able to generate a high concentration of dry hydrogen peroxide at 64 ppmv with ambient humidity at 60% RH, and air flowrate of 8.2 m/s.

## 5. Acknowledgements

This work was financially supported by the Technopolis, Suranaree University of Technology. The authors also gratefully acknowledge the support of the Synchrotron Light Research Institute (SLRI) and the Center of Excellence in Biomechanics Medicine.

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