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The Effect of Acidity and Rotation Speed in Titanium Dioxide Synthesize Process

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Abstract. The aims of this study are to analyze the effect of acidity and rotational speed in the synthesis of TiO_2 using the sol-gel method and to analyze the morphology of synthesized TiO2 nanoparticles and commercial TiO_2 using XRD to produce semiconductors for Dye-Sensitized Solar Cell (DSSC) applications. The sol-gel method was used to synthesize TiO_2 Nanoparticles. Titanium tetra-isopropoxide (TTIP) was used as a precursor with the variable of the magnetic stirrer rotation speed of 500, 1000 and 1500 rpm. Acidification was achieved by adding acetic acid to Sol-gel solution to produce a pH number of 1, 2, and 3. Nanomaterial was observed with an optical microscope and X-ray Powder Diffraction (X-RD) to determine the morphology and phase of TiO_2 crystalline. The results showed that the rotational speed and acidity level of the Sol-gel solution ware played an important role to get the best form of a nanoparticle. At a rotation speed of 1500 rpm with pH 3 and 1000 rpm with pH 2 ware shown characteristics similar to commercial TiO_2. In addition to that, the results of XRD characterization of synthesized TiO2 was shown a crystal phase of anatase structure with 18,046 nm crystal size compared to commercial TiO_2 with anatase structure and crystal size of 15,554 nm.

Introduction

The third-generation solar cell, namely solar cells made from natural Dye-sensitized solar cells (DSSCs), has attracted more attention from the researcher due to low energy and simple manufacture process[1]. DSSC is one of the potential candidates for solar cells as a source of electrical energy. DSSCs are made from abundant material so the production costs are relatively low compared to the silicon-based solar cell. DSSC light absorption and charge separation are using nano-crystal inorganic semiconductors which have a wide band gap[2]. Semiconductors are one of the four components of Dye-Sensitized Solar Cells (DSSCs) that have a role as electron carriers. Titanium dioxide (TiO_2) was the main semiconductor oxide utilize in DSSC that proven to contribute high-efficiency DSSC. TiO_2 is the heart of the DSSC itself because it was used as electron scaffold in the process to convert sunlight energy into electrical energy. The particle size of the semiconductor oxide must be on the nanoscale to enhance the overall surface area of the particles. The high surface area was contributed to increasing the amount of adsorbed dyes and the amount of light absorbed that lead to enhancement of DSSC efficiency. In addition, the semiconductor used has a porous morphology, so that the dye can enter between the pores and can be absorbed into all the surface of TiO_2 particles[3-4].

The process of synthesizing TiO_2 can be carried out in various ways including Plasma jet, hydrothermal crystallization, rapid microwave, non-hydrolytic solvothermal, and sol-gel[5-9]. In this study, the sol-gel method was used in the synthesis process since the process is easier to conduct and has advantages for each process. Compared to other methods, the sol-gel methods are widely used to control particle size and homogeneity of the TiO_2. The sol-gel method is known as one of the simple and easy methods of nanoparticle synthesis. This method is one of the "wet methods" because the process involves the solution as a medium. According to its name, the solution undergoes a phase change into sol (colloid which has suspended solids in the solution) and then becomes a gel (colloid but has a solid fraction larger than sol). The sol-gel method is suitable for the preparation of thin films and materials in the form of powder. The purpose of this preparation is for ceramic material to have a special function such as electrical, optical, magnetic, etc[10]. In this study, we proposed the effect of acidity and rotational speed to synthesized TiO_2 nanomaterial with the sol-gel method.

Methodology

The sol-gel method is one of the most successful methods of preparing nano-sized metal oxide materials. The sol is a colloidal suspension in which the dispersed phase is a solid and the dispersing phase is a liquid. The suspension of solid particles or colloidal molecules in solution, made with metal alkoxy and hydrolyzed with water, produces solid metal hydroxide particles in solution, and the reaction is called hydrolysis reaction. Gel (gelation) is a network of particles or molecules, both solids, and liquids, where polymers that occur in solution are used as a place to grow inorganic substances. Inorganic growth occurs in the gel point, where the binding energy is lower. The reaction is called condensation reaction, either alcohol or water, which produces an oxygen bridge to get metal oxide. The synthesis method using sol-gel for oxide-based materials varies depending on the precursors and the shape of the final product, whether in the form of powder, film, aerogel or fiber. The structure and physical properties of the gel depend on several things, including the selection of material raw materials, the rate of hydrolysis and condensation, and chemical modification of the sol-gel system[11]. The sol-gel method is suitable for the preparation of thin films and powder-shaped materials. The purpose of this preparation is for a ceramic material to have a special function. The sol-gel method has various benefits such as easy in composition control (homogeneous good chemical composition), low process temperature, and low cost. The precursors or starting materials in the manufacture are metal alkoxides and metal chlorides, which then undergo hydrolysis reactions and polycondensation reactions to form colloid, a system consisting of solid particles (particle size between 1 nm to 1 µm) dispersed in a solvent. The starting material can also be stored on a substrate to form a film (such as through dip-coating or spin-coating), which is then put into a container that matches the desired shape for example to produce a monolithic ceramic, glass, membranes, fiber or aerogel and to synthesize powders both micro and nano granules[12].

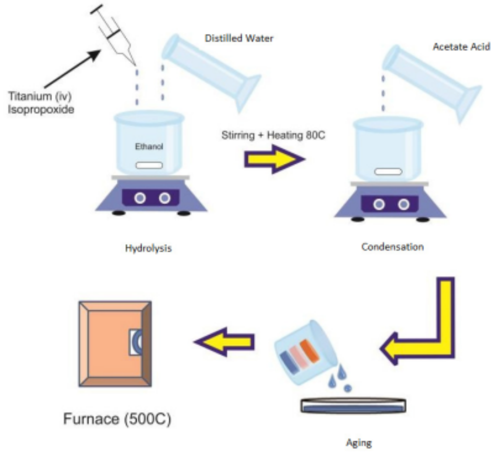


Fig. 1. Sol-Gel Process (Hydrolysis, Condensation, and Aging).

The process of synthesizing TiO_2 consists of several stages as shown in Fig. 1. The first stages were Hydrolysis, process of mixing chemicals starting with hydrolyzing the precursors or basic ingredients of TiO_2. In the process of hydrolyzing this Titanium (IV) Isopropoxide ($C_{12}H_{28}O_4Ti$), hereinafter referred to as TTIP, was mixed with ethanol and distilled water with a ratio of 1: 10: 4. Second, Condensation, The material that has been mixed was stirred using a magnetic stirrer with variations of the rotation speed of 500 RPM, 1000 RPM, and 1500 RPM at room temperature for 1 hour. The next stage of the mixture was heated with temperature 80^{o}C for 3 hours while adding acetic acid to the mixture into variations in acidity levels (pH) 1, 2, and 3. After the heating process for 3 hours, TiO_2 was changed into the form of a paste which was then left to stand for 20 hours. Third, Aging, Pasta that has been left for 20 hours was heated in a furnace (oven) with a temperature of 500^{o}C for 1 hour. This process was done in order to dry the paste, removing the solvent and the impurities.

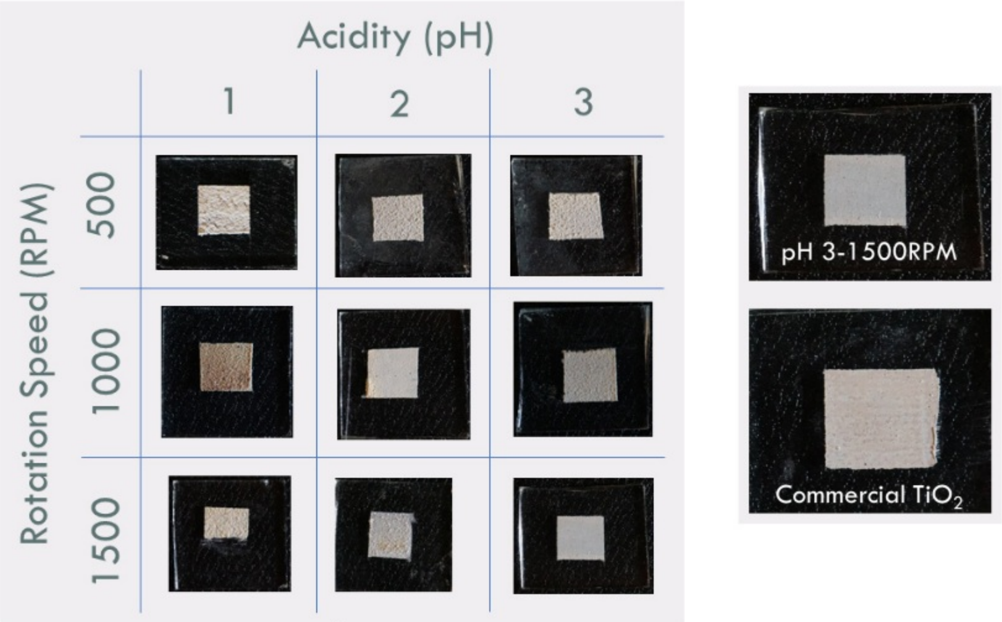
In order to analyze the final product, the sample preparation on the glass substrate was conduct as follows. First was Making Pasta, The process of making pasta begins by preparing 1 gram of hydroxyl-ethyl cellulose (HEC) stirred with 25 ml of ethanol stirred evenly until thickened. Then add 6 grams of TiO_2 (prepared by sol-gel method and commercial TiO_2 as reference). The mixture then mashed up using mortar while gradually drop 9 ml of acetic acid until it forms a paste. The second stage was, Glass Preparation, the glass that will be used as the substrate was washed with an ultrasonic cleaner. The first step was to fill the ultrasonic cleaner with water. After that, sonication was done using alternately distilled water, ethanol, and acetone. The container containing glass pieces was soaked with 15 ml of distilled water for 30 minutes. After that, it replaced with 15 ml of ethanol and sonication for 30 minutes. The last solution used was acetone with the same treatment. Third, TiO_2 Film Deposition, The next process was coating on glass with doctor blade method. The edges of the glass were framed with scotch tape, leaving a surface area of 1 cm^2 to be coated with TiO_2 paste. A deposition is done by dripping TiO_2 paste on the glass. The paste is flattened until the entire surfaces were covered with paste, then Scotch-tape can be removed. The coated substrate then heated to 500^{o}C in the furnace for 30 minutes until the layer dries (Fig. 2).

Fig. 2. Paste depositions with the doctor blade method.

The final step, X-Ray Powder Diffraction (XRD) characterization was carried out with the aim of determining the type of size, and the crystal structure of a material. The type of material can be known by comparing the results of X-RD characterization with the diffraction peak[13]. The diffraction pattern of synthesized TiO_2 nanomaterials with the sol-gel method was compared with the X-RD characterization of commercial TiO_2. TiO_2 nanoparticles were characterized using X-ray diffraction (Shimadzu X-RD-7000). The XRD results show particle diffraction patterns that are in accordance with the pattern in the Joint Committee on Powder Diffraction Standards (JCPDS).

Result and Discussion

Fig. 3 shown that pH 1 at 500 RPM the surface of the TiO_2 appears brighter compared to 1000 and 1500 RPM with the same pH. At 1000 RPM, the surface of the TiO_2 appears brownish due to the occurrence of oxidation during the synthesis process of TiO_2. Furthermore, it can be seen the comparison at pH 2 at 1000 RPM, the TiO_2 surface looks whiter and smoother than the 500 and 1500 RPM. Next is a comparison at pH 3. It can be seen that at 1500 RPM the surface of the TiO_2 appears brighter and finer than the 500 and 1000 RPM. This result shows that in order to get bright and finer surface needs a combination of acidity and rotation speed. Low rotation speed on the sol-gel process needs more acid conditions to get a finer paste. On the opposite, at high rotation speed needs less acid condition to get the best result for the TiO_2 paste.

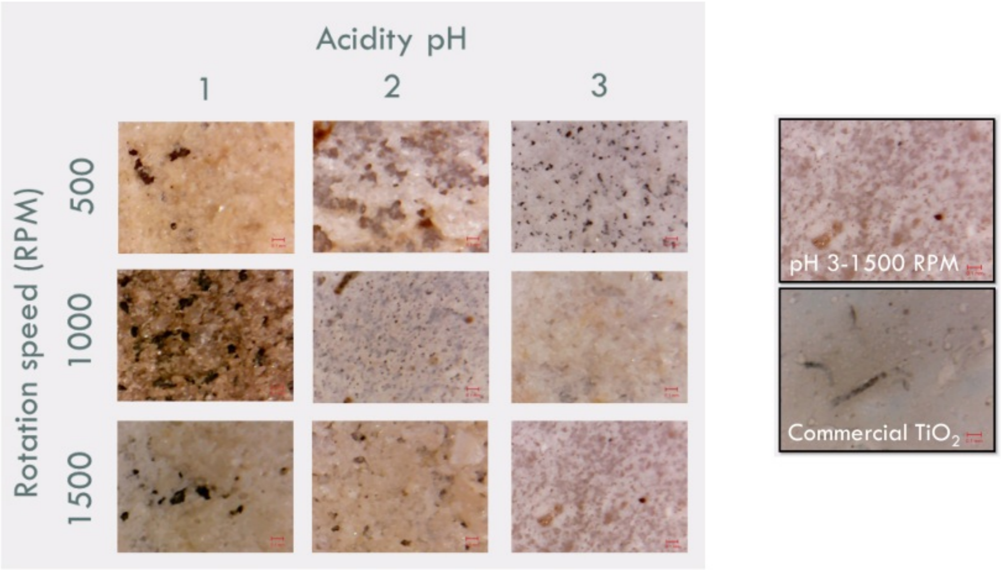


(a) (b)

Fig. 3. (a) TiO_2 film on glass substrates with acidity and rotational speed variations, (b) Comparison between synthesized TiO_2 and Commercial TiO_2.

Fig. 4 was used as a reference to determine the condition of synthesized TiO_2. The comparison image in the pH 2-1000RPM and pH 3-1500RPM looks close to the results of commercial TiO_2 of fig. 4(b). Aside from that, clearly seen some samples look rough with larger porosity. This phenomenon occurs due to the grain size was still large. In addition, the manual coating method used, the doctor blade method, made the surface thickness control was difficult to achieve. Figure 4(b) shows results with a variation of pH 3 at 1500 RPM. The surface of TiO_2 has a thin layer and porous surface even though still looks larger than commercial TiO_2. The pore with darker color and smallest dots on the surface of TiO_2 reflects the smaller pores on TiO_2 surfaces. The role of pores in titanium dioxide here plays a very important role in its use as a semiconductor in DSSC. TiO_2 which is used as a semiconductor has porous morphology so that the dye can enter between the pores and can be absorbed into TiO_2 particle surfaces. Samples with variations of pH 2 at 1000 RPM also propose the same pores appear on the surface of TiO_2 and the TiO_2 layer looks thinner. The thickness of the TiO_2 layer can also affect the efficiency of the DSSC[14]. The thickness of the TiO_2 should be optimum related to the electrolyte diffusion rate on DSSC systems[15].

Characterization by X-RD was carried out to obtain information on synthesized TiO_2 and commercial TiO_2 crystals. The synthesized TiO_2 was taken to be sampled, a variation of pH 3 at 1500 RPM. It was taken as a sample because it has a porous surface, not oxidized and has a nicer surface. The TiO_2 material analyzed was the powder taken from TiO_2 glass film after calcination. Figure 5 shows the XRD analysis of both synthesized TiO_2 and commercial TiO_2. The XRD peaks from both materials show in the same position even though clearly seen that commercial TiO_2 posed sharp peaks compared to synthesized TiO_2. The peaks provide information on the identity of the TiO_2 crystal form. The crystal shape can be known by comparing the value of 2 thetas on both diffractograms graph.



(a) (b)

Fig 4. (a) Microstructure of TiO_2 on glass substrates, (b) Comparison between synthesized TiO_2 and Commercial TiO_2.

The XRD measurement results showed that commercial TiO_2 and synthesized TiO_2 results were anatase structure which used in DSSC[16]. Through XRD diffractogram data the size of the crystal was measured by calculating the amount of FWHM (Full Width at Half Maximum). FWHM is the half-width of the diffractogram. FWHM is used to determine the size of the crystal using the Debye-Scherer equation[17]. From calculations using the Scherrer equation obtained particle diameter size of 18.046 nm for synthesized TiO_2 and 15,554 nm for commercial TiO_2.

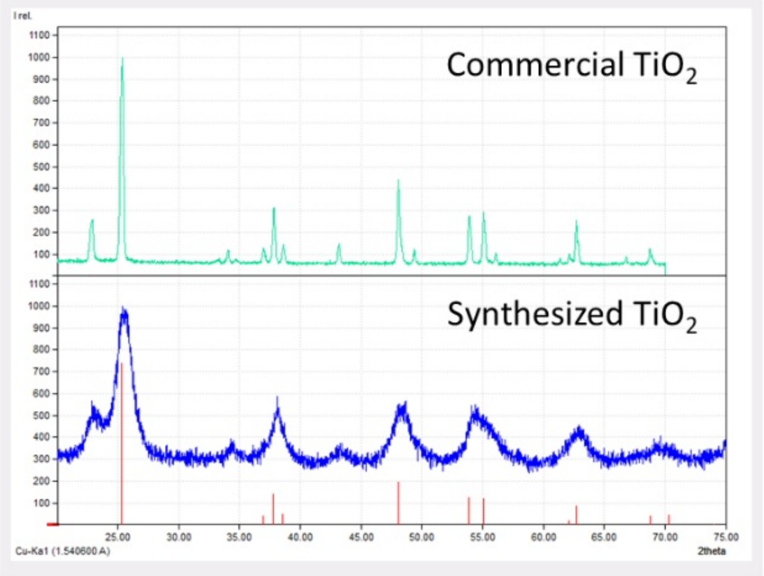


Fig. 5. XRD graphs of Synthesized TiO_2 and Commercial TiO_2.

Conclusion

In conclusion, based on the results of observations of the variations in acidity and rotational speed in the sol-gel method, it affects the quality of TiO2 film produced. At pH 3-1500RPM and pH2-1000 RPM were assessed to produce good TiO2 based on pore and surface fineness on a film substrate. It can be concluded that in less acid atmosphere a higher rotation speed was needed, and vice versa when the atmosphere is more acidic, a lower rotation speed was preferred to produce the nicer film. X-RD characteristics show the sol-gel method to synthesize titanium dioxide an anatase phase structure with a crystal size of 18,046 nm. As a comparison, commercial TiO_2 has an anatase structure with a crystal size of 15.554 nm. This data proves that the synthesized titanium dioxide with the sol-gel method can be used to manufacture titanium dioxide for DSSC application.

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