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Research Article

Physicochemical Characteristics of Titania Particles Synthesized with Gelatin as a Template Before and After Regeneration and Their Performance in Photocatalytic Methylene Blue

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Abstract

TiO_2 material has an important position in the processing of methylene blue waste because it is economical, has abundant polymorphs, high sustainability and supports green chemistry applications. Mesoporous TiO_2 is a porous material that has higher effectiveness than other TiO_2 because the pore structure has a large diameter at the nano scale (2-50 nm) with a regular shape so that the surface area and pore volume are greater than the average for other TiO_2. The synthesis of mesopore TiO_2 material has so far used the sol-gel route with synthetic pore directing agents such as P123 which can be replaced with gelatin as a cheaper and safer pore directing agent with high sustainability and abundance. Based on the description above, this research aims to photodegrade methylene blue using mesoporous TiO_2 (m-TiO_2) nanoparticles which were prepared by the sol–gel method using gelatin and P123 as template. X-ray diffraction (XRD), scanning electron microscopy (SEM), Electron dispersive X-Ray (EDX), and UV–vis spectroscopy techniques were used to characterize the samples. Photocatalytic activities of samples for methylene blue degradation were investigated. The catalyst before and after regeneration will be studied so that the effect of regeneration on the results of methylene blue photocatalysis with m-TiO_2 can be determined. XRD results confirmed the formation of the anatase and rutile phase for the TiO_2 nanoparticles, with crystallite sizes larger after regeneration in the range of 9–21 nm. The large particle size was after regeneration due to the promotion by high temperature treatment. TiO_2 nanoparticles showed the best photocatalytic activity on the first use to 91% and remained stable after four cycles with photodegradation efficiency up to 76% based on the measured UV-Vis spectroscopy. TiO_2 as synthesis could be the best candidate for catalyst with the high performance after multicycle regeneration.

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Keywords: Titania; photocatalyst; methylene blue; Physicochemical Characteristics; gelatin template

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1. Introduction

Water pollution due to dyes is considered one of the hot research topics in the contemporary world. Most industries such as textiles, paper and pulp printing, rubber, plastic, leather, cosmetics, and so on use different dyes to color their products. The total consumption of dyes in the textile industry worldwide is more than 1000 tonnes/year and around 100 tonnes/year of dyes are discharged directly into waterways [1]. The dye that is often used in the textile industry is methylene blue, which is a cationic heterocyclic aromatic compound. The complex aromatic properties with high thermal stability make it difficult to degrade in nature [2]. The threshold value for the permitted concentration of methylene blue in waters is around (5-10) mg/L [3]. The methylene blue compound has a benzene structure which is difficult to break down, is toxic, carcinogenic and mutagenic [4]. In addition, the

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rate of photosynthesis in aquatic creatures has also shown a significant decrease in the presence of toxic dyes [5]. The problem of dye waste requires great attention. Therefore, a waste processing technology is needed that can accelerate the decomposition of dye waste.

Methods for removing contaminants in waters have been studied, such as adsorption, membrane filtration, photodegradation, and biological treatment [6]. Among them, photocatalysis as a new technique has attracted attention due to its potential application in solar energy conversion as well as solving environmental problems. Because it is non-toxic, abundant, high photostability and high efficiency [7]. In photocatalytic reactions, the surface area used by the catalyst to adsorb pollutants plays an important role in the degradation rate. The advantages of the photocatalytic method are low temperature, low cost and also very low energy consumption in this method. These factors are what cause photocatalysts to be used on a commercial scale [8]. In order to be used, solar energy must be converted using photocatalytic materials, for example in the form of semiconductors. Various semiconductor materials such as TiO_2, ZnO, SnO_2, Fe_2O_3, $BiVO_4$, Cu_2O, and WO_3, have been identified as potential photocatalysts for solar energy conversion [9].

In recent years, pore nanoparticles are mainly titanium dioxide (TiO_2) in the form of anatase nanocrystals [10] has been recognized as an ideal photocatalyst for the destruction of common organic pollutants in the textile industry due to its biological and chemical inertness, stability against photochemical and chemical corrosion, electronic band gap which on photoexcitation creates oxidizing holes, and high reducing electrons [11]. Titanium dioxide (TiO_2) is an environmentally friendly material and with features of low cost and high chemical stability [6].

However, apart from having various advantages, the TiO_2 material also has several limitations, including a large band gap of around 3.2 eV. This limits its photocatalytic rate due to the rapid recombination of photogenerated electron-hole pairs. In addition, the photoefficiency is not high enough and the ultraviolet response speed is not satisfactory. Another disadvantage of pure TiO_2 material is that it only absorbs a small portion of the solar spectrum in the visible light spectrum region, thus limiting it in industrial applications because using UV lamps requires high energy [9]. Thus, the development of a photocatalytic system that can function effectively in the visible spectrum region becomes very important. Modification of TiO_2 with natural materials is of interest to many researchers to improve environmentally friendly chemical principles in the world of research.

One natural modifier is gelatin which consists of hydrophilic (polar)-hydrophobic (non-polar) groups so it can be used in the formation of porous materials such as titania [12]. Gelatin has now been widely used as a surfactant to increase the surface area of materials where the anionic functional groups of collagen play a role in improving material characteristics for optimizing thermal, physical or chemical degradation [13]. The mesoporous TiO_2 photocatalyst synthesized with gelatin is much more active when utilized in the gas phase oxidation of ethanol and acetaldehyde vapor than pure TiO_2 synthesized from P25 [14]. However, the lifetime of titania as a catalyst material plays an important role in reducing production costs and energy use. The life time of the material can be detected using the regeneration process, where ideal regeneration has the characteristics that the more regeneration cycles the degradation results do not decrease drastically, the physicochemical properties after regeneration of the material tend to be stable and it has an environmentally friendly regeneration method without the involvement of toxic reagents and minimal cost and energy.

The art of previous researches similar to this research only focused on the synthesis of mesoporous TiO_2 photocatalysts with synthetic structure-directing agents such as P123, P25, F127. A few researchers focused on use gelatin as natural source on synthesis mesoporous material. There have been limited studies concerned on on the lifetime of mesoporous TiO_2 via regeneration investigations. Based on the description above, the objectives of this research are to synthesize mesoporous TiO_2 (m-TiO_2) by sol-gel process which uses P123 and gelatin as a natural templating agent. The methylene blue photodegradation performance on TiO_2 was analyzed using the photocatalytic method using catalyst before and after regeneration. The effect of regeneration on the results of methylene blue photocatalysis with TiO_2 can be determined.

2. Materials and Methods

2.1 Materials

The ingredients used in this research were HCl 37% brand Sigma-Aldrich Merck KGaA (Mr 36.5 g/mol), commercial gelatin brand Gelita (Mr 90,000 g/mol), TEOT (Tetraethyl orthotitanate) brand Sigma-Aldrich Merck KGaA (Mr 228.109 g/mol), Pluronic P123 brand Sigma-Aldrich Merck KgaA (5800 g/mol), ethanol brand Sigma-Aldrich Merck KGaA (Mr 46.068 g/mol), and Methylene Blue brand Sigma-Aldrich Merck KGaA (Mr 319.85 g/mol), plastic wrap, label paper, and tissue.

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2.2 Methods

2.2.1 Synthesis of 1% gelatin modified mesoporous TiO_2

Mix 4 grams of pluronic triblock copolymer P123 and 1% gelatin (gelatin: P123 = 1:100(w/w) for the first variation in a beaker. Dissolve using 61mL ethanol. In another beaker, dilute 10.66 mL HCl using 69 mL distilled water. Mix the two solutions and stir the mixture using a magnetic stirrer at a temperature of 40 ºC for 1 h at a speed of 500 rpm. Then add 15.5 mL of Tetraethyl orthotitanate (TEOT) slowly and continue to stir at the same temperature and speed for 24 h in a closed condition (crepe plastic). Next, put the formed substance into a 250 mL hydrothermal autoclave and oven for 24 h at 90ºC with the bottle closed (aging stage). The resulting precipitate is filtered with filter paper until a white solid is obtained. Dry the white precipitate in the oven for 24 h at a temperature of 70 ºC. Carry out calcination at a temperature of 550 ºC for 5 h to obtain TiO_2 powder.

2.2.2 Application as photocatalyst for degradation of methylene blue

The first step in applying a photocatalyst to degrade methylene blue was to make 200 mL of methylene blue solution with a concentration of 30 ppm, by pouring 200 mL of the solution into an Erlenmeyer flask and then putting it into the photocatalytic reactor. Next, 20 mg of mesoporous TiO_2 photocatalyst was added to an Erlenmeyer containing 30 ppm methylene blue solution. The mesoporous TiO_2 adsorption process was carried out by shaking the solution in a closed, dark atmosphere in a photocatalytic reactor for 60 minutes. After adsorption for 60 minutes, the next step was to pour the methylene blue solution into 10 dark colored vials, each with a volume of 10 mL. The methylene blue solution pouring is carried out in a photocatalytic reactor in a dark atmosphere, then taking 1 vial containing methylene blue solution and labelling it as Co, which means the initial concentration before the photocatalytic process or when the photocatalytic process was still at the 0 minute stage. The other 9 vials were arranged on a shaker in the photocatalytic reactor (in the dark and without a bottle cap). The photocatalytic process began by turning on the UV lamp system (20W Xe lamp, light intensity 25 mW/cm²) with a 400 nm UV cut off filter as the light source for the degradation of methylene blue and also the shaker.

$$\%ED = \frac{C_0 - C_t}{C_0} \times 100\% \quad (1)$$

$$C_0 - C_t = kt \quad (2)$$

$$\ln\left(\frac{C_0}{C_t}\right) = -kt \quad (3)$$

where, %ED = photodegradation efficiency of MB (%), C_0 = initial concentration of MB and C_t = concentration of MB at t min.

2.2.3 Photocatalytic regeneration

The recovered catalysts (separated by filtration after the first photocatalytic cycle) were suspended in 300 mL of ultrapure water and calcinated 550 ºC for 5 h. After heat treatment process, the materials were separated by filtration, washed with water and ethanol, and dried under vacuum overnight. The dried samples were used as catalysts in the next catalytic cycle for simultaneous adsorption and discolouration of MB under UV light

2.3 Characterization instruments

The instruments used to test samples include X-Ray Diffraction (XRD) Panalytical brand (Version PW3050/60). XRD diffractogram data can be used to calculate the crystal size of mesoporous TiO_2 using the Debye Scherrer formula as in equation (4).

$$D = \frac{0.9\lambda}{B\cos\theta} \quad (4)$$

D is the crystal size in Å, λ is the wavelength used in XRD testing, namely 1.54056 Å, and B is the half-peak width in radians. θ is the angular position of the peak formation. The XRD results also show the FWHM to be able to determine the B (rad) value. Apart from knowing the phase and crystal size, the diffractogram data can also be used to calculate the crystallinity of the sample using the following formula:

$$Crystallinity = \frac{crystalline\ peak\ area}{crystalline\ and\ amorphous\ peak\ area} \times 100\% \quad (5)$$

Scanning Electron Microscopy-Energy Dispersive X-Ray (SEM-EDX) with images taken with a JEOL JSM-700 microscope at a voltage speed of 15.0 kV and Transmission Electron Microscopy (TEM). The tool for testing samples uses a Shimadzu UV-3600 Ultra Violet Visible (UV-Vis) Spectrophotometer with a wavelength of 665 nm.

3. Results and Discussion

3.1 EDX

EDX analysis aims to determine the basic composition and percentage of purity of the material contained in the m-TiO_2 sample. Figure

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1 and Table 1 show the presence of Ti and O elements in accordance with the TiO_2 compound. In the m-TiO_2 sample before photodegradation, the Ti element mapping was 46.50%, while after photodegradation it was 67.23%. The presence of gelatin which has amine groups has a high affinity to attract Ti elements so that Ti and gelatin will combine to form Ti-gelatin lumps. Furthermore, during calcination decomposition will occur in the Ti-gelatin agglomerate which causes the gelatin to be lost leaving only the Ti and O elements.

During the photocatalytic process, Ti becomes a catalytic active center that degrades methylene blue into carbon dioxide and water molecules. Then there is a process of reducing oxygen in the catalyst during regeneration where the catalyst is suspended in 300 mL of ultrapure water and calcined at 550 °C for 5 h. When heating to high temperatures, compaction occurs and the distance between atoms and Ti is reduced due to the increase in atomic packing density. Ti has a higher molecular mass than oxygen so it has higher stability than oxygen when heated at high temperatures. All the Ti elements will approach and close together, while the oxygen will partly move and also decompose with free oxygen during 550 calcination for 5 h. This is what causes the Ti component to increase after regeneration which is beneficial in the subsequent photodegradation process.

There are differences in the percentage of elements before and after photodegradation. The presence of Ti and O in EDX analysis has revealed that the synthesis of m-TiO_2 with gelatin and P123 templates was successfully carried out efficiently and at lower cost. The appearance of a

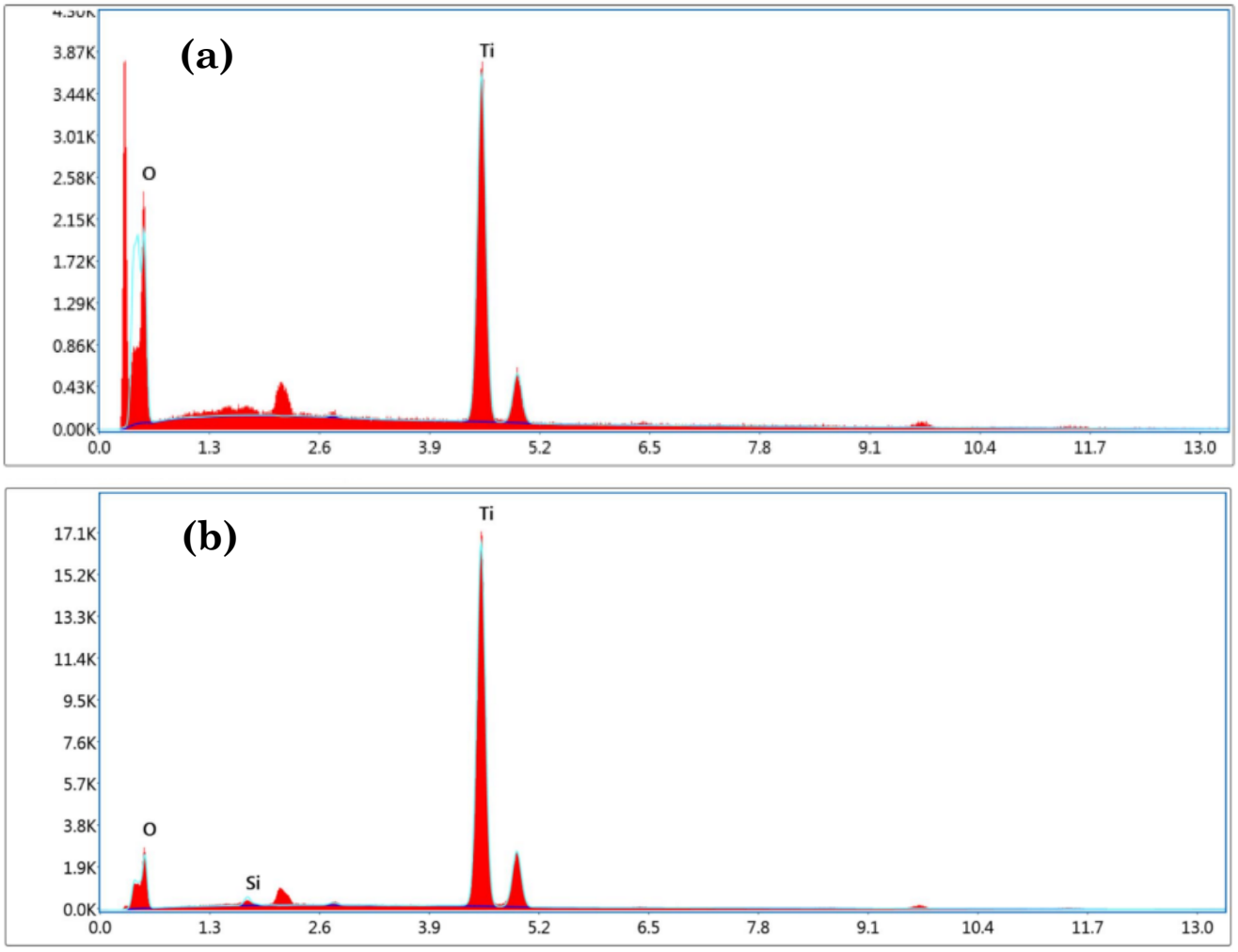

Figure 1. EDX analysis (a) before and (b) after photodegradation.

Table 1. Mapping element by EDAX from TiO_2 sample using gelatin as template.

Regeneration Sample	Mass composition, %wt		
	Ti	O	Others
Before	46.50	53.50	0
After	67.23	32.06	0.70 (Si)

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Si peak <1% after the addition of Methylene Blue dye can be considered an insignificant impurity in the photodegradation process confirms.

3.2 SEM

SEM analysis aims to see the morphology, distribution and shape of m-TiO_2 powder particles. Figure 2 shows the results of SEM characterization. Based on Figure 2, the surface morphology pattern of m-TiO_2 is an irregular shape agglomeration resulting from clumping of Ti particles between neighbors. The average size of the particles formed is 0.1 – 5.3 µm. After regeneration, it appears that the titania particles form enlarged crack rods in the size range of 14.2-20.4 µm. The large particle size in m-TiO_2 is due to the particle agglomeration process due to high temperature heating during regeneration. Where the higher the treatment temperature on the used catalyst, the more oxygen groups are pushed out and the more Ti atoms are compacted due to high temperature interactions as in Figure 2.

After regeneration, mesoporous TiO_2 appears as non-uniform aggregates that are not damaged but only enlarge due to compaction of the interatomic distances even though the porous surface of TiO_2 has been filled with methylene blue during photodegradation. This shows that the regeneration process only affects the process of increasing the degree of atomic packing and decomposition of methylene blue remaining on the titania surface due to dark adsorption.

Regeneration is one of the most important factors for assessing the quality of a catalyst material. After regenerating TiO_2 using high temperature heating, four cycles found that the m-TiO_2 morphology was well maintained due to increasing atomic packing factor.

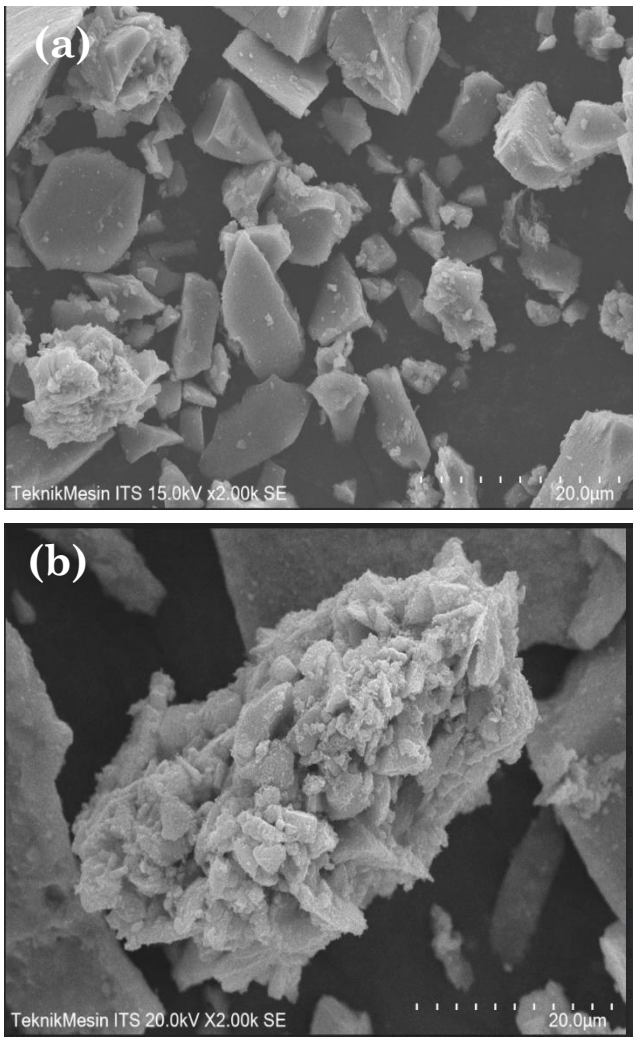

Figure 2. SEM analysis (a) before and (b) after 4 cycles of regeneration.

3.3 XRD

In this research, XRD testing was carried out. XRD testing of m-TiO_2 powder aims to determine the phase, crystallinity and crystal size of Mesoporous TiO_2 formed as a result of the synthesis that has been carried out. Based on Figure 3(a), it shows the XRD pattern of the m-TiO_2 sample before photodegradation which was prepared by calcining at 550 °C for 5 hours. The X-ray diffraction peaks around 27.4°, 36.1°, 39.1°, 41.2°, 44°, 54.3°, 56.7°, 62.7°, 64°,65.4°, 69° and 69.7° are in good agreement with the standard spectrum of rutile TiO_2 (JCPDS No. 21-1276). Meanwhile, the X-ray diffraction peaks of m-TiO_2 after degradation were at 27.3°, 36.0°, 41.1°, 43.9°, 54.2°, 56.6°, 62.6°, 64.0°, and 68.9°. Based on the X-ray diffraction peaks, it shows that TiO_2 before regeneration as fresh sample lower crystallinity and purity than after regeneration due to a decrease in order due to the accumulation of methylene blue molecules that are not completely degraded. Meanwhile, the crystallinity of TiO_2 appears to be higher due to decomposition and

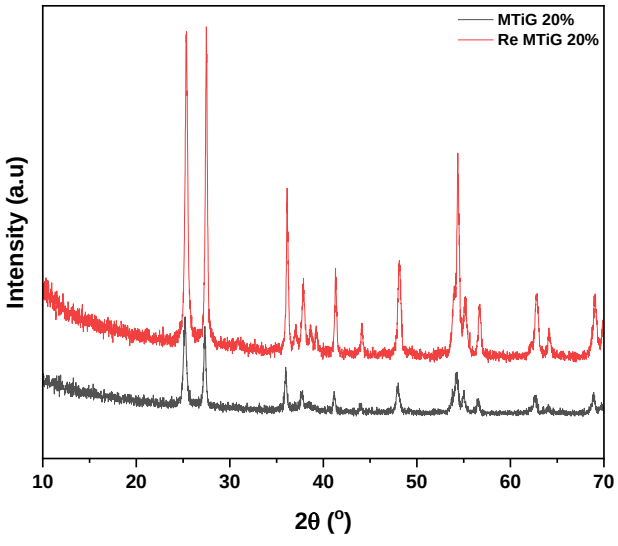

Figure 3. XRD analysis before and after regeneration.

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increase in Ti density due to repeated high temperature heating. This is in line with previous research [15].

Based on the crystal size in Table 2, it is predicted that during the regeneration process with high temperature heating, the catalyst core will be rearranged to become denser, resulting in a peak that tends to be sharp. This is because the regeneration process can increase the growth of interatomic distances by releasing weakly bound oxygen from the surface of the catalyst so that the crystallinity formed is higher and the crystal structure is more uniform [16]. Hanipa *et al* research states that the longer the heating time, the more material growth increases [17]. When viewed based on Table 2, there is a regeneration effect which increases the size of the crystals and the degree of crystallinity. In samples after regeneration, the increase in Ti particle size when reacting at high temperatures will attract other Ti elements, causing a larger crystal size.

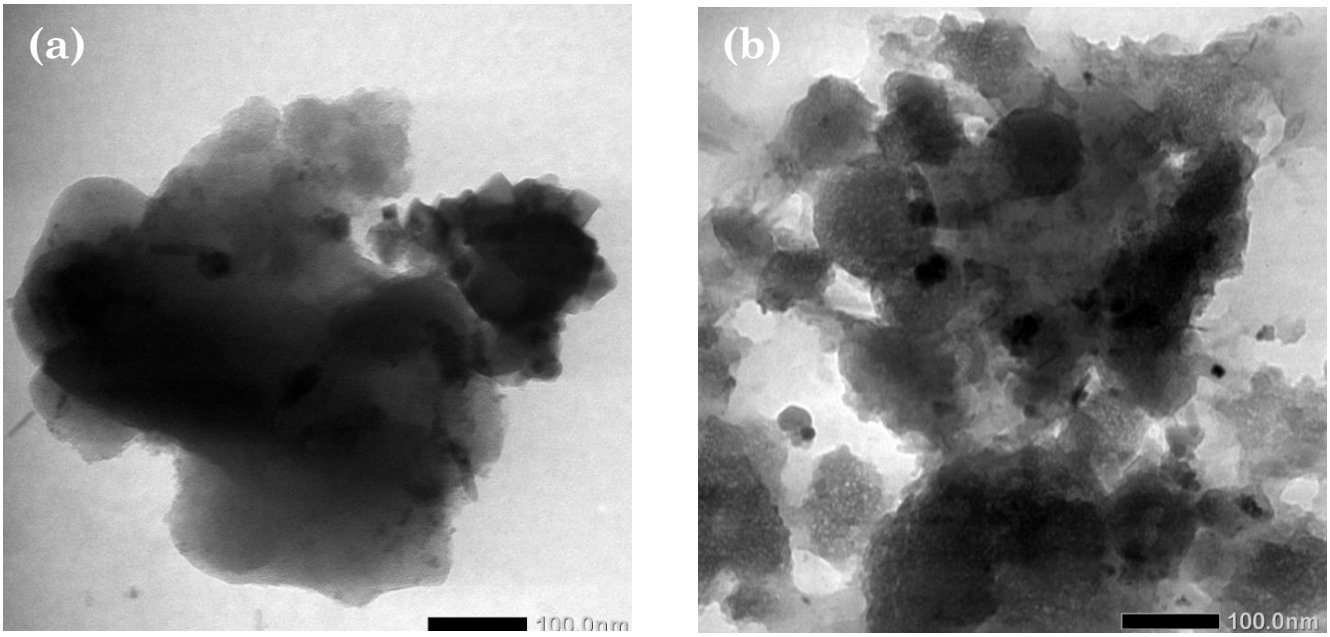

Figure 4. TEM analysis (a) before and (b) after regeneration.

Table 2. Crystal size of m-TiO_2 before and after regeneration

Pos. [°2Th.]		Height [cts]		FWHM Left [°2Th.]		*d*-spacing [Å]		Rel. Int. [%]	
Before	After	Before	After	Before	After	Before	After	Before	After
25.3662	25.2098	755.87	201.49	0.1506	0.1004	3.51131	3.53274	98.05	100.00
27.4961	27.2642	770.93	146.91	0.2007	0.1338	3.24396	3.27102	100.00	72.91
36.1292	36.0002	363.25	89.61	0.0669	0.1673	2.48618	2.49479	47.12	44.47
37.0406	-	52.91	-	0.2676		2.42707		6.86	-
37.8624	37.6205	159.92	36.68	0.2342	0.2342	2.37626	2.39099	20.74	18.19
38.6585	-	52.24		0.2007		2.32914		6.78	-
39.2703	-	47.19		0.2007		2.29426		6.12	-
41.3143	41.1318	184.35	36.65	0.2342	0.2676	2.18535	2.05816	23.91	8.05
44.0903	43.9961	53.67	16.21	0.1673	0.2342	2.05399	1.89743	6.96	27.95
48.1215	47.9459	213.09	56.31	0.3346	0.3346	1.89092	1.84978	27.64	2.49
-	49.2619		5.01	-	-	-	-	-	-
54.3668	54.2526	467.28	84.39	0.1004	0.4015	1.68754	1.69082	60.61	41.88
55.1965	55.0471	117.74	41.61	0.2342	0.3346	1.66412	1.66829	15.27	20.65
56.6427	56.5602	107.83	26.69	0.1673	0.2007	1.62502	1.62720	13.99	13.24
62.8232	62.6437	145.43	36.35	0.3346	0.2676	1.47921	1.48301	18.86	18.04
64.1343	64.0303	48.60	7.81	0.3346	0.3346	1.45210	1.45421	6.30	3.87
69.0595	-	140.82		0.2007	-	1.36008		18.27	
69.8839	68.9026	67.41	35.99	0.2676	0.3346	1.34604	1.36279	8.74	17.86
75.0613	74.9539	47.19	16.69	0.2007	0.2676	1.26552	1.26707	6.12	8.28
82.5948	-	41.09	-	0.8029	-	1.16814		5.33	-
84.3257	-	16.32	-	0.4015	-	1.14853		2.12	-

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XRD diffraction patterns before and after regeneration, no changes can be observed in the crystal structure of m-TiO_2 with similar diffraction patterns which are in line with previous studies [18]. The relatively high photodegradation results after 4 cycles of regeneration on m-TiO_2 indicate that m-TiO_2 has good stability, supported by the XRD pattern after adsorption which does not show significant changes as in Figure 3 diffraction peaks at 2θ = 25.3º (002), 36.1º (311), and 62.8º (440) can still be observed.

3.4 TEM

TEM analysis was carried out on 1% gelatin modified titania samples. This characterization is carried out to see the particle size of the sample. The results of the morphological analysis and particle size distribution are shown in Figure 4. The results of the TEM analysis in Figure 4 show that until the worm hole-like m-TiO_2 produced, there are certain parts that are triangular or square-like in shape, originating from the representation of the macroscopic shapes of anatase and rutile. By measuring the grain size using ImageJ software from several particles, the average particle size for triangle-like mesopores with a diameter of 5-9 nm was produced.

TEM analysis of mesoporous silica in Figure 4(b) after regeneration shows the formation of irregular worm-hole like with size 20-30 nm due to high temperature heating which decomposes the remaining methyl blue on the surface as well as oxygen in the Ti framework due to compaction between Ti atoms. The triangle-like structure disappears but the worm hole like character increases consisting of irregular pores similar to pebbles. Similar things were also found in previous research regarding the effects of high heating on materials [19].

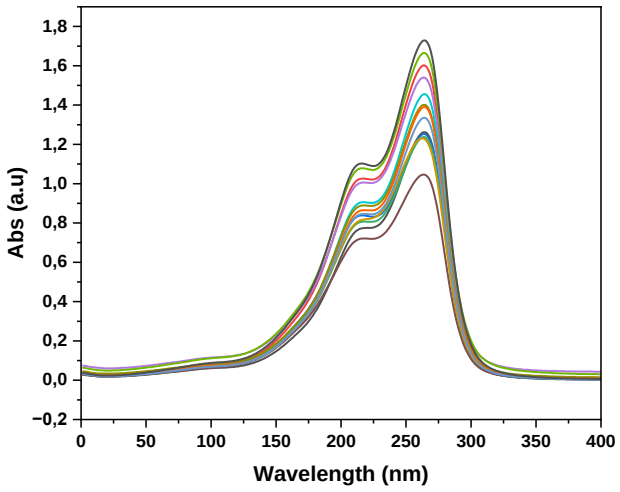

Figure 5. Methylene Blue after Photocatalytic treatment Cycle 4

3.5 Photocatalytic Regeneration

Figure 5 show the photocatalytic result which is show that methylene blue is degraded by fresh sample in zero cycle and sample after regereneration in first to fourth cycle (Figure 5). Methylene blue dark adsorption by each sample reached zero efficiency in 60 min but after 60 min of UV irradiation, the concentration of the methylene blue solution had decreased obviously. The rate for photocatalytic degradation of methylene blue by fresh m-TiO_2 was the fastest. The time required for photocatalytic degradation of methylene blue in fresh m-TiO_2 which the final catalytic efficiency reached 91.0% was shorter than after first regeneration cycle for MB. The first factor for high photodegradation performance is crystallinity. Based on the X-ray diffraction peaks, fresh m-TiO_2 has lower crystallinity which is influence to enhance both of surface area. After regeneration, not only decrease surface area but also increase crystal size of Ti due to the attract betweena Ti elements as result of high temperature reaction.

The second factor is decrease active sites after regeneration as TEM explanation in Figure 4. The formation of irregular worm-hole like with size 20-30 nm after regeneration is larger than before regeneration due to Ti atoms compaction as high temperature result (Figure 2). The decrease of oxygen after Ti compaction decrease the functional group number for interact with methylene blue (Table 1 and Figure 1). The result of this factor in decrease photodegradation performance after regeneration.

The regeneration and reuse potential of mesoporous silica was determined using thermal treatment and chemical desorption with solvents. Desorption experiments were carried out after completion of the adsorption process by thoroughly washing the composite with ultrapure water to destroy the electrostatic interaction between methylene blue and m-TiO_2. After the m-TiO_2 with methylene blue which was not degraded was separated from the solution through filtration then dried at a temperature of 60 ºC and calcined at a temperature of 550 ºC for 5 h then the photodegradation process was repeated 4 times with the same procedure.

The results of the cyclic experiment in Figure 6 show that the percentage of methylene blue photodegradation reached 88.83% in the first cycle and decreased to 71.64% after the fourth cycle. Despite its reduced efficiency, mesoporous TiO_2 still shows good potential as a reusable catalyst for removing methylene blue from wastewater. The decrease in the percentage of methylene blue photodegradation after each cycle can be caused by particle damage and compaction of Ti so that it is estimated that there is a decrease

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in the number of pores. This causes a decrease in the contact area between the catalyst and the methylene blue molecules, thus reducing the overall active sites of the m-TiO_2 composite. In general, the regeneration results show that the TiO_2 material synthesized from gelatin and P123 has good stability and regeneration performance, making it a sustainable catalyst for wastewater treatment.

The quality of the catalyst before and after regeneration can be seen from the XRD diffraction pattern before and after methylene blue photodegradation (Figure 3) which shows no peak shift, indicating that the catalyst process has high stability. The same thing can also be seen from the SEM analysis results (Figure 2 which shows that the morphological structure of the material did not experience significant damage after regeneration. Changes in catalyst elements can be seen from the EDX analysis results showing that m-TiO_2 after photodegradation has 0.70% Si and 32.06% O. The high oxygen concentration in m-TiO_2 before regeneration provides a greater possibility of interaction between the oxygen atoms in silica and the hydrogen atoms in methylene blue, to increase the adsorption capacity. However, this reduction in oxygen still provides a photocatalytic efficiency of 70-80% after cycle 4 which indicates that the life time of the catalyst is quite large and is capable of being an economical catalyst with a good level of structural stability.

4. Conclusions

The mesoporous TiO_2 synthesis process was successfully carried out using the soft template method with TEOT precursor and gelatin and P123 surfactants using the hydrothermal sol-gel method. The result is a photocatalyst with a fine white powder texture. The sample had a degradation efficiency value (%) of 91.18%, then after 4 cycles of regeneration the degradation efficiency value became 71.64%. The results of investigations into the character of the TiO_2 catalyst synthesized using gelatin-P123 before and after regeneration in methylene blue photodegradation shows no change in crystallinity except changes in intensity due to compaction of the Ti element and decomposition of oxygen with methylene blue at high temperatures. Compaction of Ti elements in the catalyst after regeneration increases the Ti content in the catalyst based on EDX results. SEM analysis shows that the morphological structure of the synthesized TiO_2 experiences an increase in size due to repeated heating with an irregular wormhole-like shape resulting from 4 cycles of regeneration, indicating that the synthesized TiO_2 catalyst has high stability after high temperature heating.

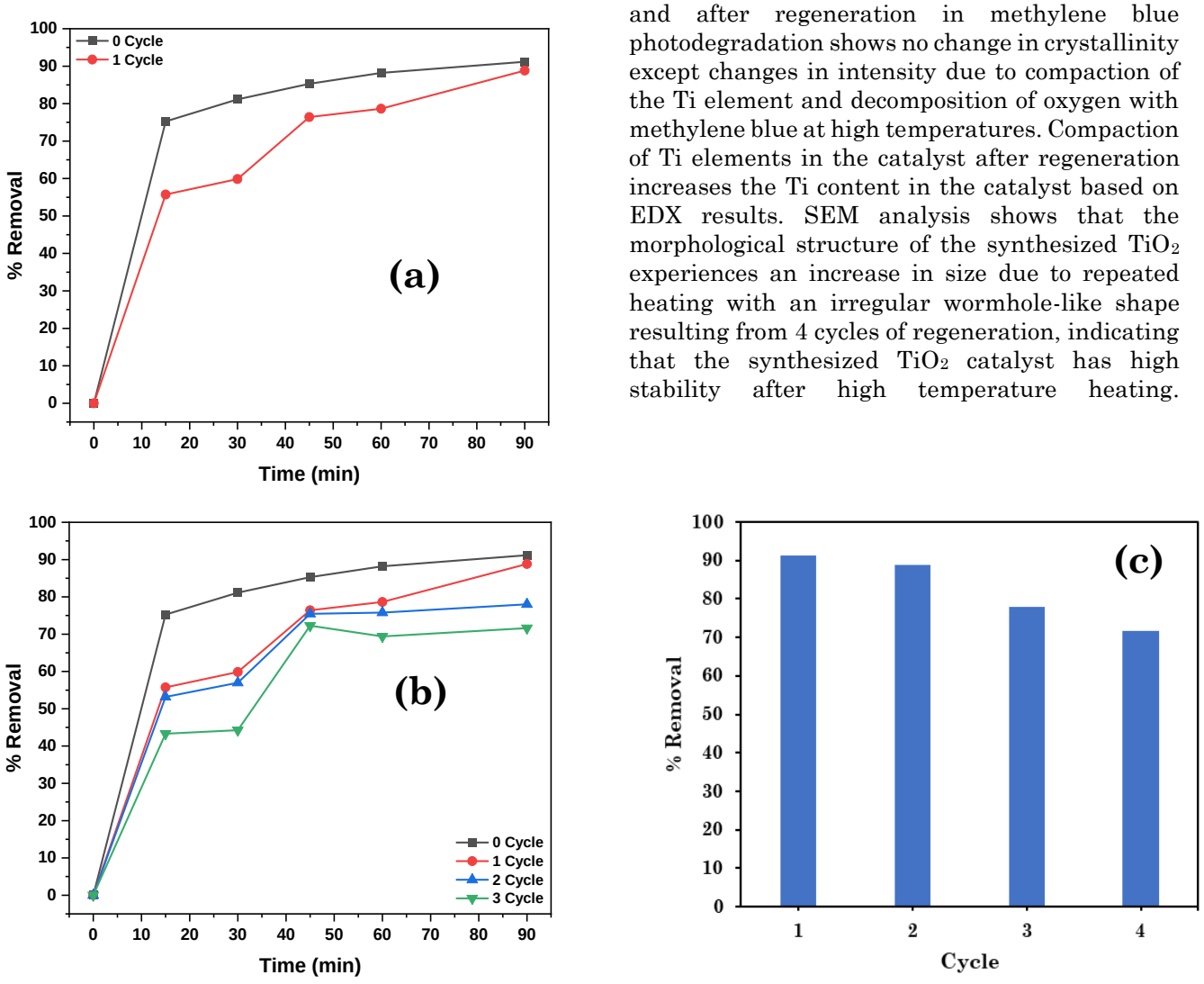

Figure 6. Diagram of % Removal m-TiO_2 at 4 cycles

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Therefore, it can be concluded that this photocatalyst has the potential to become a future photocatalyst candidate because it has high regeneration power.

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CRediT Author Statement

Author Contributions: *M. Ulfa:* Conceptualization, Investigation, Resources, Data Curation, Writing, Review and Editing, Supervision; *M. Ulfa and I. Pangestuti*: Methodology, Formal Analysis, Data Curation, Writing Draft Preparation; *M. Ulfa and C. N Anggreani:* Review and Editing, Data Curation, and Validation. All authors have read and agreed to the published version of the manuscript.

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