Effect of microwave power intensity on synthesis of mesoporous titania nanoparticles for degradation of 2,4-dichlorophenol: Photoactivity performance and kinetic studies

N A Marfur, N F Jaafar* and F H H Gani

School of Chemical Sciences, Universiti Sains Malaysia, 11800 USM Penang, Malaysia

* nurfarhana@usm.my

Abstract. In this study, mesoporous titania nanoparticles (MTN) were synthesized via microwave (MW)-assisted method under various heating power (0.12 (MTN1), 0.37 (MTN2) and 0.56 (MTN3) W g⁻¹). The performance of MTN was investigated on photocatalytic degradation of 2,4-dichlorophenol (2,4-DCP) in aqueous solution using a batch reactor under visible light irradiation. The catalysts were characterized by XRD, UV-Vis DRS, BET and FTIR. The degradation of 2,4-DCP showed that catalyst synthesized at MTN2 was the most effective with 89.43% degradation compared to MTN1 and MTN3 with 82.93% and 86.99%, respectively. This result probably MTN2 has the lowest band gap and highest surface area compared to MTN1 and MTN3. The kinetic studies for degradation 2,4-DCP using MTN2 revealed that the reaction followed pseudo-first order kinetics with reaction rate values (k_{app}) decreased as the initial concentration increased that proved the system was preferable at low concentration.

1. Introduction

Studies for the environmental betterment have been a huge concern of many parties for recent decades in order to preserve the environment for a longer period. Influx of poisonous untreated discharge produced mainly in industrial sectors contribute in environmental pollutions [1]. For instance, chlorinated compounds or other organic compound are often not easily degradable and exposure to them may result in severe diseases [2].

Chlorophenols (CPs) including 2,4-dichlorophenol (2,4-DCP) are pollutants that draw researchers’ attention due to the high risk of harming the environment and human health [3]. They are broadly used in industrial activities such as insecticides, herbicides and wood preservatives where large consume of these substances introduces more harmful contaminants to nature [4]. To remove these adverse pollutants from ecosystem, many have attempted various efforts such as wet oxidative, immobilization and oxidation degradation but the methods possess several disadvantages such as time and cost ineffective as well as produce large amounts of by-products [5].

Advanced Oxidation Processes (AOP) is a method which capable of producing reactive oxygen species (ROS) such as hydroxyl radicals (•OH) under proper light irradiation and particular catalyst [6]. AOPs have become a favourable method as they can transform large amount of pollutants at a time into a more durable and harmless compounds [7]. Photocatalysis is one of the AOPs and can be explained as a chemical reaction that is driven by light absorption (usually ultraviolet or visible light) of solid substance (or photocatalyst) that undergo no chemical changes during and after the reaction [8].

Numerous semiconductors have been used in this field such as TiO₂, ZnO, SnO and ZnS [9]. Among them, TiO₂ has been a comprehensive catalyst that are extensively used in photocatalysis due to its potential of being potent and stable under light exposure [10]. Research suggest that TiO₂ is
suitable for the environment due to its properties of inertness, chemically and thermally, as well as high stability to light irradiation and chemical corrosion [11]. However, the drawbacks of TiO$_2$ has limited its efficiency. Currently, continuous studies related to mesoporous transition metal oxide have been conducted since it can improve its initial properties. Mesoporous titania has been intensely studies due to its miscellaneous implementations [12]. Therefore, alteration in titania morphology can resolve its drawbacks by generating numerous site defects and lowering of band gap [13].

Microwave (MW)-assisted method is one of the potential approaches to synthesis mesoporous material by assisting nucleation, gives better heat distribution and reduces the reaction period and particle size compared to conventional heating method [14-15]. Study shows MW method can produce catalyst with large surface area, pore volume, pore width as well as enhancing the crystal growth [16]. Microwave power density is an important parameter for determination of crystal structure and surface area including reduce the particle size along with band gap while generating many site defects which contribute in hindering electron-hole from recombine and aiding charge carrier migration. Research by Jaafar et al. (2015) demonstrated that mesoporous titania nanoparticles (MTN) catalyst prepared by sol-gel method followed by MW heating did a good performance on the photocatalytic degradation of 2-chlorophenol [17].

In this study, different MTN were synthesized via MW approach under various heating power ranging from 0.12 to 0.56 W g$^{-1}$ and applied for photocatalytic degradation of 2,4-DCP under visible light irradiation. Besides that, other parameters that could affect the performance of the catalyst in this research have been studied including pH of the pollutant, concentration of the pollutant and the catalyst dosage. The kinetic analysis of the best catalyst was also studied for more understanding related to the interaction of catalyst and pollutant during the photocatalytic degradation process according to Langmuir-Hinshelwood model.

2. Experimental

2.1. Reagents, materials and apparatus
Titanium (IV) isopropoxide (TTIP) was bought from Sigma-Aldrich. Cetritrimethyl-ammonium bromide (CTAB) surfactant, propanol and hydrochloric acid (HCl) were purchased from MERCK, Malaysia. Acetone was purchased from HmbG Chemical and methanol was purchased from RPE Reagent pure Erba. Sodium hydroxide (NaOH) and ammonium hydroxide (NH$_4$OH) were purchased from QREC™ and 2,4-DCP from Alfa Aesar, Germany with 99% purity.

2.2. Preparation of catalyst
Mesoporous titania nanoparticles (MTN) were synthesize using microwave (MW)-assisted method. 4.68g of CTAB was dissolved in 720 mL of distilled water, 120 mL of propanol and 29 mL of 28% ammonia solution. The mixture then stirred continuously for 30 min at 323 K in water bath. After 30 min, the temperature of the water was increased to 353 K and followed by addition of 5.7 mL of TTIP and continued stirring for 2 h in water bath. After 2 h, the white solution was transferred to a 1000 mL beaker and placed in microwave for heating (Samsung ME711K) which the power can be operated in the range of 100-800 W and frequency of 2.45 GHz. The heating was done sporadically continued for 2 h in order to attain a sol-gel form of MTN. The power of the microwave was varied with 100, 300 and 450 W denoted as MTN1, MTN2 and MTN3, respectively. The product was collected and dried overnight in oven followed by calcination at 873 K for 3 h.

2.3. Characterization of the catalysts
A Bruker Advance D8 X-Ray powder diffractometer (XRD) with Cu K$\alpha$ radiation ($\lambda=1.5418$ Å) at 20 angle ranging from 15° to 75° was used to identify the crystalline structures of the catalysts. The phases were identified with the assist of the Joint Committee on Powder Diffraction Standard (JCPDS) files. UV-Vis DRS (Perkin Elmer Lambda 900) spectrophotometer was used to measure the diffuse reflectance of the catalyst in air at room temperature with the wavelength ranging between 200-500
nm. Fourier Transfer Infrared (FTIR) spectroscopy (Perkin Elmer Spectrum 2000 FT-IR) was used to identify the chemical functional groups present in the catalysts. IR absorbance data were obtained between the range of wavenumbers from 400 to 4000 cm⁻¹. Brunauer-Emmett-Teller (BET) method was used to identify the surface area, pore distribution and isotherm.

2.4. Photocatalysis degradation of 2,4-dichlorophenol
Photocatalytic degradation of 2,4-dichlorophenol (2,4-DCP) was performed to test the photoactivity of each catalyst. The photocatalytic experiments were carried out in a batch reactor equipped with fluorescence lamp (48 Watt) which was used as the source of light. 0.0750 g of catalyst was added in 200 mL of 2,4-DCP solution. The solution was stirred in dark for 2 h to achieve adsorption desorption equilibrium followed by 6 h of reaction under light irradiation under continuous stirring. The initial pH of the solution used was 5 and the reaction was carried out at 303 K. The concentration of the 2,4-DCP in the solution prior to irradiation was used as the initial value for the 2,4-DCP degradation measurement.

Aliquots of 2 mL of the solution was taken during the reaction with time interval of 30 min and placed in conical centrifuge tubes. The solutions are centrifuged in a Hettich Zentrifugen Micro 120 at 45 000 rpm for 15 min before being analyzed by UV-Vis spectrophotometry (Shimadzu Corp, UV-2600 UV-Vis Spectrophotometry) for the residual concentration of 2,4-DCP. Each set of photocatalytic activity experiments are repeated thrice to ensure the accuracy of the experiment. The adsorption band of 2,4-DCP was taken at 284 nm and the degradation percentage was calculated using the equation below:

\[
\text{Degradation} \% = \frac{C_0 - C_t}{C_0} \times 100\% \quad (1)
\]

where \(C_0\) and \(C_t\) are the initial concentration and the concentration at time of 2,4-DCP, respectively.

3. Results and discussion

3.1. Characterization of the catalyst

3.1.1. X-ray diffraction. Figure 1 shows the diffraction peaks of the catalyst prepared under various MW watt. A series of XRD peaks of TiO₂ anatase was observed (JCPDS file no. 00-004-0477) with peaks at 25.32° (101), 37.14° (200), 37.50° (004), 38.23° (112), 48.15° (200), 54.27° (105), 55.33° (211) and 62.51° (204). The peak intensity of MTN has an increasing pattern proportional to the power density from 100 W to 450 W. The particle size of MTN1, MTN2 and MTN3 are 13.25, 12.24, 11.36 nm, respectively, which were calculate using Debye-Scherrer equation:

\[
D = \frac{k\lambda}{\beta \cos \theta} \quad (2)
\]

where \(D\) is the particle size, \(\lambda\) is the wavelength of the X-ray radiation (Cu Kα =0.1542 nm), \(k\) is the shape factor (\(k =0.94\)), \(\beta\) is the line width at half-maximum height, and \(\theta\) is the angular position of the peak maximum, \(2\theta = 25.30°\). The pattern for peak intensity is in increasing order from the lowest to the highest MW power where Figure 1(c) shows highest intensity. It can be deduced that it has the highest crystallinity due to good structural arrangement of TiO₂ during synthesis that contributed by appropriate heat distribution by MW.
3.1.2. FTIR. The catalysts were subjected to FTIR analysis and the spectra are shown in Figure 2. According to figure 2A, all catalysts exhibited a moderate sharp band at 3541 cm\(^{-1}\) which are attributed to hydroxyl group (OH). Figure 2B shows all catalysts have a band at 1626 cm\(^{-1}\) that are assigned to OH vibration of the surface-adsorbed water [18]. Bands at 1025 cm\(^{-1}\) (MTN1 and MTN2), 1150 cm\(^{-1}\) (MTN3) and at 450 cm\(^{-1}\) for all catalysts are assigned to the presence of Ti-O-Ti asymmetrical stretching and vibrational mode [19]. According to Bezrodna (2004), a wide band at 550 cm\(^{-1}\) attributed to Ti-O-Ti vibration was observed for the commercial TiO\(_2\) [20]. In this study, the particular band is slightly shifted to the right at 450 cm\(^{-1}\) could be due to the microwave heating that gives different strength of Ti-O bonds caused by oxygen adsorption.

3.1.3. BET. Figure 3A shows the nitrogen adsorption-desorption of the catalysts. All catalysts exhibited isotherm type IV with a H3 hysteresis loop that proved a typical adsorption profile for meso-structured material with slit-shaped pores that are non-uniform in size or shape [21]. The hysteresis loop at P/P\(_0\) = 0.65-0.99 was assigned to nitrogen condensation occurs within neighbouring nanoparticle voids that were formed due to textural porosity between particles [22]. In spite of the fact that adsorption trends are almost similar for all catalysts but there is better nitrogen adsorption in the region for MTN2 compared to MTN1 and MTN3. This recommends that the surface area and pore structure might be influenced due to difference in microwave power [23]. The BET surface area for MTN1, MTN2 and MTN3 are 68.36, 116.8 and 73.18 m\(^2\)/g, respectively. The higher BET surface of MTN2 compared to other catalysts might the main reason for the higher photocatalytic activity reported in this study.

Figure 3B demonstrates the pore distribution for all catalysts in the range of 1.5 to 36 nm. The catalysts were seen to possess larger pore sizes in the range of 23-34 nm that suggests the pore formation are favourably occurred in the latter compared to the former TiO\(_2\). The average pore sizes of 22 nm for the MTN2 were slightly smaller than that of MTN1 and MTN3. Larger crystallites aggregation and merging of the pores could be seen in MTN2 compared to the remaining two catalysts.
Figure 2. (A) FTIR spectra in region 4000-2215 cm\(^{-1}\) and (B) FTIR spectra in region 2000-425 cm\(^{-1}\) of (a) MTN1 (b) MTN2 and (c) MTN3.

3.1.4. **UV-Vis DRS.** The UV-Vis DRS spectra of the catalysts are shown in Figure 4. All catalysts showed single sharp edges with the band-gap absorption onset at 360.0, 378.0 and 369.0 nm, respectively. The band gap energies obtained were calculated using the following equation:

\[
eV = \frac{hc}{\lambda} \quad (3)
\]

where \(h\) is Planck’s constant \((6.626 \times 10^{-3} \text{ m}^2 \text{ kg/s})\), \(c\) is speed of light \((3.000 \times 10^8 \text{ m/s})\) and \(\lambda\) is the wavelength values corresponding to the intersection point of the vertical and horizontal part of the spectra. The values of band gap for MTN1, MTN2 and MTN3 are 3.420, 3.280 and 3.370 eV, respectively. Many previous studies related to the narrowing of the band gap of catalyst as a main contribution for a better photocatalytic activity.

3.2. **Photocatalytic activity of the catalyst**

3.2.1. **Effect of microwave heating.** Based on the graph (Figure 5), it can be seen that microwave heating at 300 W (MTN2) has a more stable increase in percentage of degradation than the remaining two. The total percentage of degradation of MTN2 was 89.43% while MTN1 and MTN3 gave 82.93% and 86.99% respectively. However, this contradicted with previous research by Jaafar (2015) that reported the highest degradation of 2-chlorophenol (2-CP) with 96.00% using MTN synthesised at 450 W [16]. In this study, MTN2 has shown the highest efficiency in the degradation might be due to
the highest surface area and the lowest band gap of the catalyst as reported. MTN2 has the smallest band-gap value and this supported by the result of the degradation of the 2,4-DCP.

Figure 3. (A) N$_2$ adsorption-desorption isotherm and (B) pore distribution of (a) MTN1 (b) MTN2 (c) MTN3.

Figure 4. UV-vis spectra of (A) MTN1, (B) MTN2 and (C) MTN3. (Extrapolation straight line: band gap energy value)
3.2.2. Effect of pH. The pH of the solution was one of the significant parameters in analysing the photocatalytic reaction. The series of pH used were 3, 5, 7, 9 and 11. Based on the results obtained (Figure 6A), the best degradation was obtained at pH 5 with 89.43% whereas at pH 3, 7, 9 and 11 were 88.62%, 87.73%, 80.95% and 80.95%, respectively. The result is in agreement with Figure 6B, where shows the amphoteric properties of MTN2 contributes to the result that was obtained. This can be elaborated using amphoteric properties of the catalyst which depends on its Point of Zero Charge (PZC) [22]. The PZC of the MTN2 was found to be pH 6.4 as shown in the graph. Any pH higher than this value will demonstrate that the surface of the catalyst is negatively charged and conversely [1]. The catalyst surface in acidic solution predicted to be positively charged while negatively charged in a more alkaline solution [24].

Strong electrostatic field that existed in between the organic materials of the pollutant that carried negative charges and MTN2 that carried positive charges influenced the activity of the catalyst [25]. This corresponds to the previous studies that proposed surface of TiO$_2$ were positively charge at low pH value while 2,4-DCP were negatively and neutrally charged. Thus, lower pH values enhanced the adsorption of the pollutant for better degradation [26].

3.2.3. Effect of concentration. Optimization was furthered with varying the initial concentration of 2,4-DCP in the range of 10-100 mg L$^{-1}$ using MTN2. Figure 7A shows that at the lowest concentration (10 mg L$^{-1}$), the rate of degradation was the most promising with 89.43% indicated that penetration of light on the surface of catalyst were high during degradation. As the initial concentration of 2,4-DCP are increased, the degradation decreased gradually. This is much likely because increase in initial concentration with unchanged dosage, light intensity and duration could restrict and reduce the light penetration to the surface of the catalyst where the formation of hydroxyl is reduced too which plays important role in the photocatalytic degradation [25]. Hence, the available OH radicals are inadequate for the pollutant degradation at higher concentrations. As a result, the pollutant degradation rate
decreases as the concentration increases [27]. The kinetic for 2,4-DCP degradation using MTN2 was studied based on Langmuir-Hinshelwood model [25]. Figure 7B exhibits the linear graph of ln (C₀/Cₜ) against irradiation time. From the graph, it can be concluded that the photodegradation obeyed pseudo-first order kinetics models [28]. Based on Table 1, decrease in kₚ value as concentration increase also indicated that the system worked better at low concentration [29]. The calculated kᵣ (reaction rate constant) and Kᵢₗ (adsorption coefficient) values were 0.4075 mg L⁻¹ min⁻¹ and 0.0179 L mg⁻¹, respectively indicating that 2,4-DCP was the controlling step during the reaction [30].

![Graph showing pH vs. Degradation](image)

**Figure 6.** (A) Effect of pH on MTN2 during degradation of 2,4-DCP and (B) isoelectric point (pH₉₉⁰) of MTN2. (C₂₄DCP = 10 mg L⁻¹, W = 0.075 g L⁻¹, t = 8 h, T = 303 K).

**Table 1.** Percentage degradation at different initial concentrations of 2,4-DCP and pseudo-first order apparent constant values for 2,4-DCP degradation using MTN2. (pH=5, W=0.075 g L⁻¹, t = 8 h, T=303K).

| Initial 2,4-DCP concentration, C₀ | Degradation (%) | Reaction rate, kₚ (x10⁻³ min⁻¹) | Initial reaction rate, r₀ (x10⁻² mg L⁻¹ min⁻¹) |
|-----------------------------------|-----------------|---------------------------------|-----------------------------------------------|
| 10                                | 89              | 6.2                             | 6.2                                           |
| 30                                | 87              | 4.7                             | 14.1                                          |
| 50                                | 86              | 4.1                             | 20.5                                          |
| 70                                | 80              | 3.0                             | 21.0                                          |
| 100                               | 79              | 2.7                             | 27.0                                          |

3.2.4. Effect of dosage. The photodegradation was carried out with various catalyst dosage of 0.0190, 0.0375, 0.0750, 0.0940 and 0.1130 g. As observed in Figure 8, the highest degradation was with catalyst dosage of 0.0375 g. As the catalyst dose were increased from 0.0190 g to 0.0375 g, a slight increase in degradation rate was observed due to the presence of high number in surface active sites on
the catalyst surface. However, further increase of catalyst dose decreased the degradation rate. The rate of degradation started to decrease might be caused by the low number of surface active sites on the catalyst surface due to aggregation [31]. Furthermore, amount of light penetration to the 2,4-DCP at high concentration might be decreased due to the turbidity of suspension in the solution [32].

3.2.5. Proposed mechanism of the study. Based on the experimental finding, a mechanism with schematic energy level diagram is proposed to represent the charge transfer process for the degradation of 2,4-DCP as shown in Figure 9. When MTN2 is irradiated by visible light, the electron in the valence band (V_B) of TiO_2 can be excited to its conduction band (C_B), forming an electron-hole pair (4). Since the band-gap of the MTN2 is the lowest, the easier to induce the electron transfer from V_B to C_B.

\[
\text{TiO}_2 \xrightarrow{hv} \text{TiO}_2 + \text{h}_\text{V_B}^+ + \text{e}_\text{C_B}^- \quad (4)
\]

The hole then combines with H_2O to form •OH radicals (5) and the electrons reduces O_2 to superoxide anion radicals, •O_2^- (6).

\[
\begin{align*}
\text{H}_2\text{O} + \text{h}_\text{V_B}^+ & \rightarrow \cdot\text{OH} + \text{H}^+ \\
\text{O}_2 + \text{e}_\text{C_B}^- & \rightarrow \cdot\text{O}_2^- 
\end{align*} \quad (5) \quad (6)
\]

The excited electrons fully generate •OH (7) to mineralize 2,4-DCP partially or completely (8).

\[
\begin{align*}
\cdot\text{O}_2^- + \text{H}^+ & \rightarrow \cdot\text{HO}_2^- \rightarrow \text{OH}^- \\
\cdot\text{OH}^- + 2,4\text{-DCP} & \rightarrow \text{degraded 2,4-DCP} 
\end{align*} \quad (7) \quad (8)
\]

**Figure 7.** (A) Effect of concentration on MTN2 during degradation of 2,4-DCP and (B) photodegradation kinetic of 2,4-DCP using MTN2 at different initial concentration (a) 10 mg L^{-1}, (b) 30 mg L^{-1}, (c) 50 mg L^{-1}, (d) 70 mg L^{-1} and (e) 100 mg L^{-1}. (pH=5, W=0.075 g L^{-1}, t = 8 h, T=303 K).
Figure 8. (A) Effect of catalyst dosage on MTN2 during degradation of 2,4-DCP. ($C_{2,4-DCP} = 10$ mg L$^{-1}$, pH=5, $t = 8$ h, $T$=303 K).

Figure 9. The proposed mechanism for the degradation of 2,4-DCP.
4. Conclusion
In this study, mesoporous titania nanoparticles were successfully synthesized using simple microwave-assisted method. XRD results shows that the intensity of peaks for TiO$_2$ increased as the microwave heating power increased while the surface area found from BET analysis revealed that MTN2 has the highest surface area compared to the remaining two and directly becoming the main contribution for its good performance in degrading 2,4-DCP. Moreover, the lowest band-gap is also observed with MTN2 which could be mainly contributed for it to be the most efficient catalyst in this study. Although the band-gap values for the catalysts are higher than previously reported values, the degradation of the pollutant are sufficiently good.

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