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Polluted River Water Treatment Via Pilot-Scale Membrane Photocatalytic Reactor (MPR) Incorporated Zno-Kaolin Under Different Light Intensity

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Abstract

Polluted river water treatment utilizes a mix of physical, chemical, and biological processes and activities. Conventional systems, including coagulation, flocculation, sedimentation, filtration, and disinfection, have several limitations. Hence, the Membrane Photocatalytic Reactor (MPR) is one of the most promising methods for polluted river water treatment. ZnO-Kaolin nanoparticles served as great photocatalysts for MPR performance. This study focuses on the pilot-scale hybrid MPR treatment of polluted river water under different light intensities (100, 125, and 225 watts) and irradiation times (20, 30, and 40 minutes). The treated water quality analysis is based on the Environmental Quality Act 1974 (EQA 1974). The kinetic rate was also investigated using pseudo-first-order and pseudo-second-order models. It was found that 225 watts and 30 minutes were the optimum values. The pilot scale hybrid MPR proved to fit well with the pseudo-second-order kinetic models, suggesting that the degradation follows a chemisorption mechanism. In conclusion, we believe the hybrid MPR pilot-scale system will enhance efficiency in cleaning dirty river water, all while meeting the standards of the EQA 1974.

1. Introduction

River water is frequently polluted with agricultural, industrial, and human refuse, which poses an undeniable threat to human health [10]. Micropollutants, such as endocrine-disrupting compounds (EDCs), and infectious

microorganisms, such as viruses and pathogenic bacteria, are significant contributors to this problem [10], [14]. Currently, river water treatment utilizes a mix of physical, chemical, and biological processes and activities. There are several stages in this conventional system, which are coagulation, flocculation, sedimentation, filtration, and disinfection. However, this conventional system has several limitations. One of the limitations is that the resultant sludge is a byproduct of any form of coagulation operation. Recently, literature reported that MPR systems have great performance in water and wastewater treatment. The membrane photocatalytic reactor (MPR), a novel hybrid approach for water and wastewater treatment that combines photocatalysis with a membrane separation mechanism, is a new hybrid method for water and wastewater treatment. Nontoxicity and continuous operation are two of the system's advantages. Previous research claimed that the laboratory scale of MPR has the best performance for polluted water treatment and produces the highest quality of permeate. However, the performance of pilot-scale MPR for polluted river water treatment has not been well discovered. Recently, it was confirmed that ZnO nanoparticles have great performance in MPR for polluted water treatment. However, the performance of ZnO-Kaolin in MPR has not been confirmed. Furthermore, the effect of irradiation light on the photocatalytic performance of different semiconductors must be investigated since it may lay the foundation for a better knowledge of photocatalysis principles.

The significance of this study is to present and discuss the advanced utilities and equipment currently used in the industry for effluent treatment. This study examines the influence of light intensity and retention time on the performance of MPR. This endeavor also includes the development and formulation of a kinetic study for this research project. Meanwhile, this study focuses on the following aims in order to treat polluted river water via pilot-scale MPR incorporating ZnO-Kaolin under different light intensities and irradiation time. Next, to compare and analyze before and after the quality of treated water. Lastly, to establish the kinetic reaction model of photocatalytic degradation for production's scaling up. In the expected outcomes of this study, the pilot-scale hybrid MPR is believed to attain better efficiency to be elucidated for polluted river water and comply with the Environmental Quality Act of 1974. In addition, it is expected that the kinetic reaction rate model of photocatalytic degradation for pilot-scale hybrid MPR will be successfully established and elucidated according to pseudo-first order and pseudo-second-order models.

2. Material and Method

This section summarizes the experimental work that has been performed, which is the treatment of polluted rivers via different light intensities and irradiation times. Secondly, the analysis of the quality of treated water and the analysis of the kinetic reaction rate model of photocatalytic degradation.

2.1 Material

The material and apparatus that used in the experiments are filter paper, measuring cylinder 10 mL and 100 mL, analytical balance, UV lamp, pH meter, pH paper, beakers 500mL and 1000mL, dissolve oxygen meter, turbidity meter, RO water, distilled water, water bath, filtering flask and filter funnel. The chemical components utilised in the experiment are 0.1M Sodium Hydroxide (NaOH) (Sigma Aldrich, US), 0.1M Hydrochloric acid (HCl) (Sigma Aldrich, US), Kaolin Powder (Sigma Aldrich, US) and vial digestion solution for COD.

2.2 Pilot Scale MPR Water Treatment

The sampling of 80 liters of river water was collected from the Sembrong River's. It is collected by wading into the midpoint of the river and facing in the direction of the flow. Before the experiment, the ZnO-Kaolin was weighed using a lab scaler at a catalyst concentration of 0.05 mg/L loading. Next, we added 80L of polluted river water to the feed tank to mix the weighted ZnO. The ZnO-Kaolin solution was recirculated for approximately 30 minutes to achieve a uniform catalyst concentration. After 30 minutes of mixing, the solution was transferred to the photocatalytic reactor using a submersible pump. Inside the photocatalytic reactor, the mixture was circulated for intervals of 20, 30, and 40 minutes while exposed to aeration and a UV lamp interval of 100, 125, and 225 watts. Next, the UV lamp and aeration were both turned off at the same time to stop the photocatalyst reaction in the reactor. Lastly, the photocatalyst reactor's solution is transferred to the membrane module, which is then circulated for around 10 minutes. A sample of 150 mL was taken at the membrane module as permeate.

2.3 Water Quality Analysis

Based on Fig. 1, the Biochemical Oxygen Demand (BOD) was estimated by first determining the dissolved oxygen (DO) of the treated water. The treated water DO was determined using a DO meter (Hach). Prior to that, the buffer solution was created. The pH of the samples was tested and changed to achieve the standard pH of 7, using Sodium Hydroxide and Hydrochloric Acid solutions. The DO of the samples was measured using a DO meter (Hach) on the first day. Following that, samples of treated water with a defined quantity of oxygen were stored in an incubator at 20 °C for five days to determine BOD5. Next, the COD of the permeate was measured using a *DR*6000™ UV-VIS

Spectrophotometer (Hach) using the usual protocol supplied by Hach. The permeate samples were first pre-heated to 150 °C in a DRB 2000 reactor. Futhermore, the AZ 86031 was used to determine the concentration of DO when the probe was put into the sample water. Move the probe into the water to expel any air bubbles and deliver a new sample to the sensor cap. Moreover, the turbidity of treated water was measured using a Hach turbidimeter and the normal protocol supplied by Hach. The data was read in Nephelometric Turbidity Units (NTU). Next, we used a Hanna instrument pH meter to measure the pH of the permeate, following the Hanna instrument standard protocol. After that, the ammonia nitrogen test was measured using a $DR6000^{TM}$ UV-VIS spectrophotometer (Hach) based on the Nessler method. All result analysis needs to be compared with Environment Quality Analysis 1978 (EQA 1978). Finally, the reductions in BOD, COD, turbidity, and ammonia nitrogen were calculated using the formula below:

$$\text{Reduction} = \left[\left(\frac{C_0 - C_f}{C_0}\right) \times C_0\right] \times 100\% \tag{1}$$

where, C_0 is the initial concentration of respective analysis and C_f represents the final concentration.

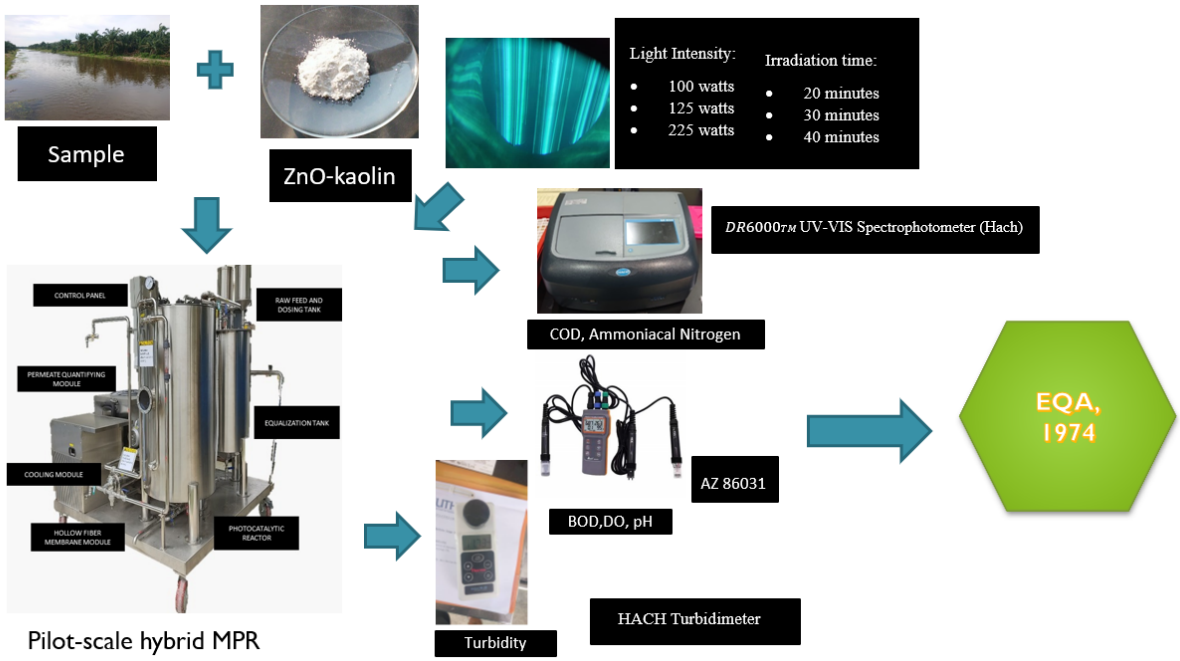

Fig. 1 *Methodology of treating polluted river water*

2.4 Kinetic-Study of Photocatalytic Degradation

The kinetics study was determined using two models which are pseudo-first Order Model and Pseudo-Second Model, as in Eq. (2) and (3), respectively.

$$\ln(C_e - C_t) = \ln C_e - K_t \tag{2}$$

$$\frac{t}{C_t} = \frac{1}{KC_e^2} + \frac{t}{C_e} \tag{3}$$

where, C_e is the concentration at equilibrium, C_t represent the concentration at time, t, and K_t is the rate constant. A graph of $\ln(C_e - C_t)$ vs. t was constructed for Eq. (2), while a graph of t/C_t vs. t was constructed for Eq. (3). The slope was used to determine the rate constant, K, and later was used to determine the photocatalytic degradation reaction rate.

3. Result and Discussion

This result and discussion summarize the experimental work performed utilizing the technique given in the material and methodology in terms of data collection, analysis, and conclusions. Furthermore, the findings and outcomes of the study were compared to previous studies described in the literature review.

3.1 Characteristic of Untreated River Water

Table 1 below shows that the EQA 1974 and water quality units were acquired from Malaysia's National Water Quality Standards (NWQS). The paper is used as a reference to compare the water quality standards before and after treatment.

Table 1 *Analysis of untreated water and treated (blank water sample)*

Parameters	*Units*	*EQA 1974*	*Untreated polluted river water*	*Treated polluted river water**
BOD	mg/L	1	12.76	-
COD	mg/L	10	54.89	38
DO	mg/L	7	6.19	7.6
Turbidity	NTU	5	36.13	36.8
pH	-	7	4.67	6.28
Ammoniacal Nitrogen	mg/L	0.1	2.26	1.26

*A "blank water sample" is a sample that does not contain the analyte of interest in analytical chemistry.

3.2 Correlation of Light Intensity and Irradition Time Pilot-Scale Membrane Photocatalytic Reactor

The graph of the data experiments according to the parameter used to determine the water quality is shown in Fig. 2 (a). The data parameters collected from the photocatalytic reactor were BOD, COD, DO, turbidity, pH, and ammoniacal nitrogen. We collected the sample water after processing it for 30 minutes at 100 watts, 125 watts, and 225 watts in the photocatalytic reactor. Figure 4.1 shows the correlation between light intensity and the quality of treated water. First, the results of an experiment that has been done to see the effect of UV-light power using different powers of UV-light (100, 125, and 225 watts) on the reduction of Biochemical Oxygen Demand (BOD). Before treatment, the BOD was 12.76 mg/L. At 100 watts, the BOD reduction was 99.72%. Next, at 125 watts, the BOD reduction was 97.89%. Lastly, at 225 watts, the BOD reduction is 99.38%. The power of UV light and Biochemical Oxygen Demand (BOD) have an inverse relationship: as UV light power rises, BOD reduction increases. This implies that stronger UV radiation may decrease BOD in water more efficiently. Next, the results of an experiment that has been done to see the effect of UV-light power using different powers of UV-light (100, 125, and 225 watts) on reduction of Chemical Oxygen Demand (COD). Before treatment, the COD was 54.89 mg/L. At 100 watts, the COD reduction was 74.49%. Next, at 125 watts, the COD reduction was 89.07%. Lastly, at 225 watts, the COD reduction is 90.89%. This indicates that as the intensity of UV light increases, the level of reduction in Chemical Oxygen Demand (COD) increases. This phenomenon occurs because UV light has the ability to decompose substances in water, leading to a reduction in the oxygen required for oxidizing these substances [13]. Chemical oxygen demand (COD) measures the quantity of oxygen needed to oxidize all organic materials within water. Furthermore, the results of an experiment that has been done to see the effect of UV-light power using different powers of UV-light (100, 125, and 225 watts) on Dissolved Oxygen (DO). Before treatment, the DO was 6.16 mg/L. At 100 watts, the DO was 6.90 mg/L. Next, at 125 watts, the DO was 7.73 mg/L. Lastly, at 225 watts, the DO is 6.20 mg/L. The concentration of dissolved oxygen (DO) decreases or increases as UV light power increases. This is due to the ability of UV radiation to decompose dissolved oxygen into oxygen atoms. Photolysis is the process of breaking down dissolved oxygen using UV radiation [3].

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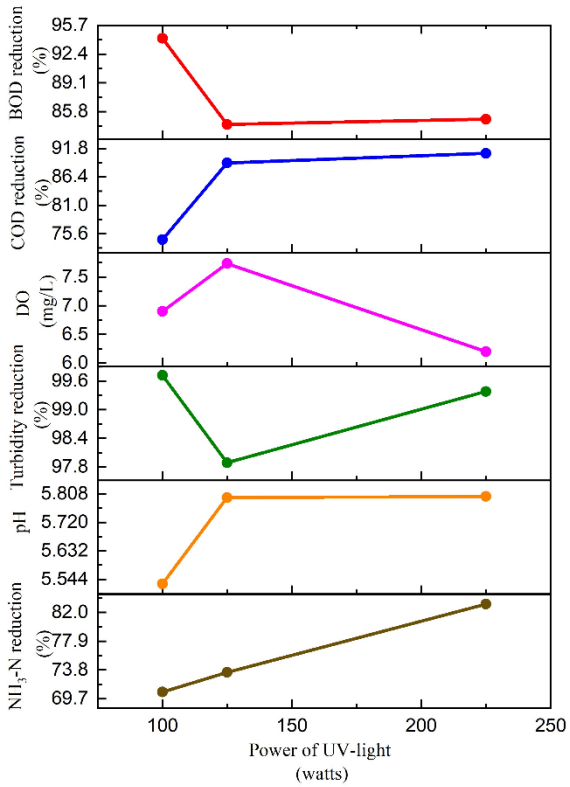

Fig. 2 *(a) Correlation of light intensity at analysis quality of treated river water*

After that, the results of an experiment that has been done to see the effect of UV-light power using different powers of UV-light (100, 125, and 225 watts) on the reduction of turbidity. Before treatment, the turbidity was 36.13 mg/L. At 100 watts, the turbidity reduction was 99.72%. Next, at 125 watts, the turbidity reduction was 97.89%. Lastly, at 225 watts, the turbidity reduction is 99.38%. The reduction in turbidity increases as UV light power increases. This is because UV radiation may break down the particles that create turbidity, such as suspended solids, bacteria, and algae. Photolysis is the breakdown of particles by UV light. Then, the results of an experiment that has been done to see the effect of UV-light power using different powers of UV-light (100, 125, and 225 watts) on pH. Before treatment, the pH was 4.67. At 100 watts, the pH was 5.53. Next, at 125 watts, the pH was 5.80. Finally, at 225 watts, the pH is 5.80. This shows that the higher the power of UV-light, the higher the pH in a membrane photocatalytic reactor due to the direct effects of UV-light on water as well as the formation of hydroxyl radicals. At last, the results of an experiment that has been done to see the effect of UV-light power using different powers of UV-light (100, 125, and 225 watts) on the reduction of ammonia-nitrogen. Before treatment, the ammonia nitrogen concentration was 2.26 mg/L. At 100 watts, the ammonia reduction was 70.65%. Next, at 125 watts, the ammonia-nitrogen reduction was 73.45%. Lastly, at 225 watts, the ammonia-nitrogen reduction is 83.19%. The reduction of ammonia nitrogen increases as the intensity of UV light increases. This is due to the ability of UV light to break down ammonia into nitrogen and hydrogen. In conclusion, we have selected 225 watts of light intensity in the photocatalyst as the optimal value, as each analysis of treated polluted material demonstrates a higher effective value in accordance with the Environmental Quality Act of 1974's standards.

The graph of the data experiments according to the parameters used to determine the water quality is shown in Fig. 2(b). The data parameters collected from the photocatalytic reactor were BOD, COD, DO, turbidity, pH, and ammoniacal nitrogen. We collected the sample water after processing it for 20 minutes, 30 minutes, and 40 minutes at 225 watts in the photocatalytic reactor. First, the results of an experiment that has been done to study the effect of photocatalytic time contact using different times of UV light (20, 30, and 40 minutes) on the reduction of Biochemical Oxygen Demand (BOD). Before treatment, the BOD was 12.76 mg/L. In 20 minutes, the BOD reduction was 90.83%. Next, at 30 minutes, the BOD reduction was 84.93%. Lastly, at 40 minutes, the BOD reduction is 89.97%. This indicates that as the photocatalytic time of contact increases, the level of reduction in Biochemical Oxygen Demand (BOD) increases. Thus, longer photocatalytic time contact often results in greater BOD elimination effectiveness. Next, move to the results of an experiment that has been done to study the effect of photocatalytic time contact using different times of UV light (20, 30, and 40 minutes) on the reduction of chemical oxygen demand (COD). Before treatment, the COD was 54.89 mg/L. After 20 minutes, the COD reduction was 98.18%. Next, at 30 minutes, the COD reduction was 90.89%. Lastly, at 40 minutes, the COD reduction is 89.07%. This illustrates that as the photocatalytic time of contact increases, the level of reduction in chemical oxygen demand (COD) increases. Therefore, the longer a pollutant is exposed to the photocatalyst under UV

irradiation, the longer it has to undergo degradation. Furthermore, the value of dissolved oxygen (DO) is based on the effect of photocatalytic time contact using different times of UV light (20, 30, and 40 minutes). Before treatment, the value of DO was 6.16 mg/L. At 20 minutes, the value of DO was 7.57 mg/L. Next, at 30 minutes, the value of DO was 6.20 mg/L. Lastly, at 40 minutes, the value of DO is 7.70 mg/L. This shows that as the photocatalytic time of contact increases, the level of dissolved oxygen (DO) increases. Thus, the duration of photocatalytic contact time is the amount of time that sample-polluted river water is exposed to an active photocatalyst under UV light.

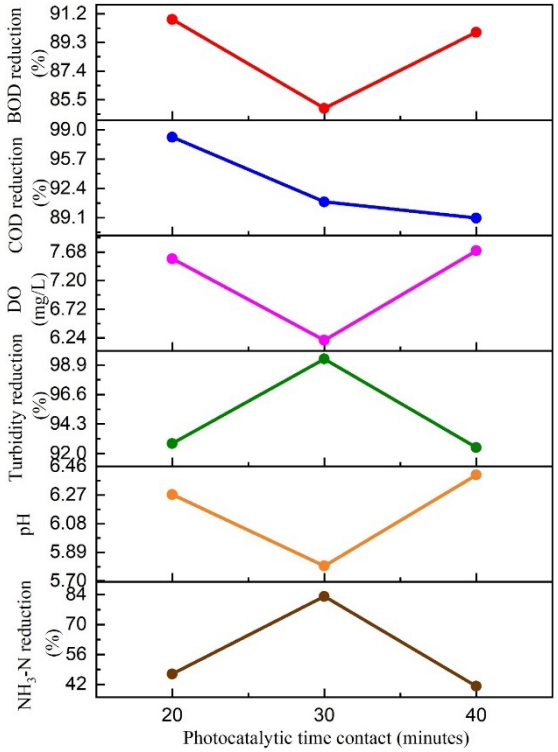

Fig. 2 *(b) Correlation of irradiation time at analysis quality of treated river water*

Subsequently, the results of an experiment were done to study the effect of photocatalytic time contact using different times of UV light (20, 30, and 40 minutes) on the reduction of turbidity. Before treatment, the turbidity was 36.13 mg/L. In 20 minutes, the turbidity reduction was 92.79%. Next, at 30 minutes, the turbidity reduction was 84.93%. Lastly, at 40 minutes, the turbidity reduction is 89.97%. This shows that as the photocatalytic time of contact increases, the level reduction of turbidity increases. The duration of photocatalytic contact time refers to the duration of exposure of sample-polluted river water to an active photocatalyst under UV light. Next, the value of pH is based on the effect of photocatalytic time contact using different times of UV light (20, 30, and 40 minutes). Before treatment, the pH value was 4.67. At 20 minutes, the value of pH was 6.27. Next, at 30 minutes, the pH value was 6.20. Lastly, at 40 minutes, the pH value is 6.40. This shows that as the photocatalytic time of contact increases, the level of pH increases. This is because longer photocatalytic contact durations give more possibilities for UV light to activate the photocatalyst [7]. At last, the results of an experiment that has been done to study the effect of photocatalytic time contact using different times of UV light (20, 30, and 40 minutes) on the reduction of ammoniacal nitrogen. Before treatment, the ammonia nitrogen concentration was 2.26 mg/L. In 20 minutes, the ammonia-nitrogen reduction was 92.79%. Next, at 30 minutes, the ammonia-nitrogen reduction was 99.38%. Lastly, at 40 minutes, the ammonia nitrogen reduction is 92.47%. This indicates that as the time of the photocatalyst increases, the level of reduction of ammoniacal nitrogen increases. This is because longer photocatalytic contact durations allow for more extensive UV light activation of the photocatalyst. In conclusion, we have selected 30 minutes of photocatalytic time contact as the optimal value, as each analysis of treated polluted material demonstrates a higher effective value in accordance with the standards of the Environmental Quality Act of 1974. Additionally, the connection between photocatalytic time contacts and the result of the analysis of a polluted river is not linear. There is a threshold for diminishing returns when extending the photocatalytic time contact beyond a certain point does not result in a meaningful increase in the values of analysis, treatment, or elimination.

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3.3 Analysis Kinetic Reaction Model of Photocatalytic Degradation

From the graph pseudo-first-order model, it was indicated that there was a relationship between $\ln(q_e - q_t)$ and times in minutes in Fig. 3(a), and from the graph pseudo-second-order model, it was indicated that there was a relationship between t/q_t and times in minutes in Fig. 3(b). The power of UV-light consists of three powers, which are 100 watts, 125 watts, and 225 watts. The graph was plotted to forecast the $\ln(q_e - q_t)$ if the time is added from 30 minutes to 210 minutes. From this graph, the linear graph equation was generated in Table 4. In this study, two kinetic models were developed based on the experimental data to investigate their applicability. Fig. 3(a) showed that pseudo-first-order kinetic models have the lowest linear regression (R^2) values, which proved that the models were not suitable and inapplicable. However, as shown in Fig. 3(b), the values (R^2) for pseudo-second-order kinetic models were the highest. Therefore, it clearly proved that the pseudo-second-order kinetic models for photocatalytic degradation were applicable, suggesting that the degradation may follow chemisorption. Chemisorption occurs when the molecule of synthetically polluted river water is held on the ZnO-Kaolin surface by chemical forces such as short covalent chemical bonding through the sharing of electrons [8].

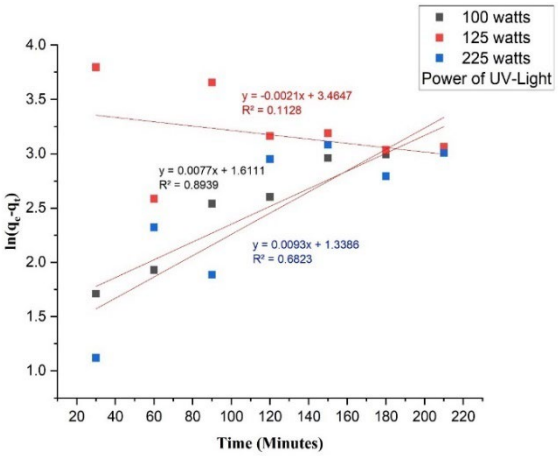

Fig. 3 (a) *The relationship between ln qe-qt*

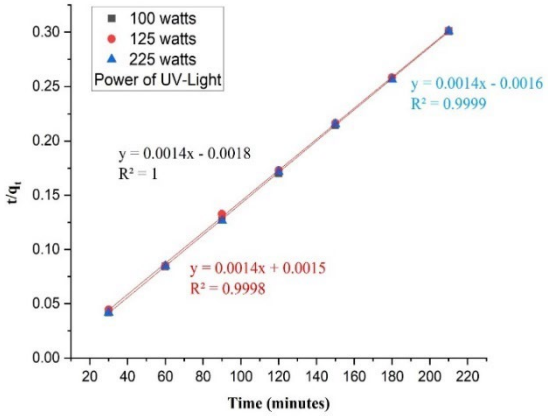

Fig. 3 (b) *The relationship between* t/q_t *and times in minutes*

From this graph, the linear graph equation was generated in Table 4. In this study, two kinetic models were developed based on the experimental data to investigate their applicability. From Table 4, it clearly showed that pseudo-first-order kinetic models have the lowest linear regression (R^2) values, which proved that the models were not suitable and inapplicable. However, from Table 4, the values (R^2) for pseudo-second-order kinetic models were the highest. Therefore, it clearly proved that the pseudo-second-order kinetic models for photocatalytic degradation were applicable, suggesting that the degradation may follow chemisorption. Chemisorption occurs when the molecule of synthetic polluted river water is held on the ZnO-Kaolin surface by chemical forces as short covalent chemical bonding through the sharing of electron [8].

Table 4 *Linear graph equation of pseudo-first-order and pseudo-second-order*

Power of UV-light (Watts)	Pseudo-first-order		Pseudo-second-order	
	Equation	Radiant, R^2	Equation	Radiant, R^2
100	$(y = -0.0021x + 3.4647)$	0.1128	$(y = 0.0014x - 0.0018)$	1.000
125	$(y = -0.0077x + 1.6111)$	0.8939	$(y = -0.0014x + 0.0015)$	0.9998
225	$(y = -0.0093x + 1.3386)$	0.6823	$(y = -0.0014x - 0.0016)$	0.9999

4. Conclusion

To conclude, the percent of turbidity removed from river water is 99.38% at 225 watts and 30 minutes in photocatalytic time contact, as compared to the National Water Quality Standards (NWQS), proving that the polluted river water has been successfully treated. As a result, the project's objective, which investigated the light intensity and photocatalytic time contact loading optimum values in a pilot plant Membrane Photocatalysts Reactor (MPR), was met. According to the experiment's findings, the ideal power of UV light is 225 watts and 30 minutes in the photocatalytic reactor. The 225 watts were used as light intensity to help achieve this outcome because of their advantages in terms of high stability and high photosensitivity. The power of UV light (225 watts) assists river water treatment by speeding up chemical processes. Thus, the experiment revealed that a major

factor in ensuring the catalyst's ability to react with water is the concentration of the photocatalyst. Furthermore, the 30 minutes were used as photocatalytic time contact to help achieve this outcome because of its advantages in terms of high stability, high photosensitivity, and lower cost. The photocatalytic time contact (30 minutes) assists river water treatment by speeding up chemical processes.

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Conflict of Interest

The authors declare that there is no conflict of interest regarding the publication of the paper.

Author Contribution

The authors confirm their contribution to the paper as follows: ***study conception and design:*** *Amirrul Hakim Suhaimi, Rais Hanizam Madon, Nur Hanis Hayati Hairom;* ***data collection:*** *Mohammad Alif Hakimi Hamdan, Amirrul Hakim Suhaimi;* ***analysis and interpretation of results:*** *Rais Hanizam Madon, Nur Hanis Hayati Hairom, Siti Nur Fatin Nadhirah Mohd Makhtar;* ***draft manuscript preparation:*** *Amirrul Hakim Suhaimi, Zuliazura Mohd Salleh, Mzahar Abd. Jalal, Rais Mohd Hazri Madon. All authors reviewed the results and approved the final version of the manuscript.*

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