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RESEARCH ARTICLE

EVALUATION OF THE SYNERGISTIC EFFECT IN WASTEWATER TREATMENT FROM SHIPS BY THE ADVANCED COMBINATION SYSTEM

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ABSTRACT

The shipping industry accounts for 90% of global trade. The remarkable growth of the shipping sector over the past decade has created many environmental challenges. The discharge of wastewater from ships (containing black and gray water) has become a serious threat to the marine environment. Developing a real wastewater treatment system not only meets the requirement of small space onboard but also meets the latest standards of the International Maritime Organization (IMO) remains a challenging task for the shipping industry. To remove oil, anionic surfactants, and organic substances in domestic wastewater, ship bilge water, treatment methods combining advanced oxidation and adsorption processes were applied in this study. In this paper, a combined system including ozonation and photocatalysis combined with adsorption on activated carbon to ensure the simultaneous removal and decomposition of organic contaminants in polluted wastewater from ships. The influence of system operating parameters on the efficiency of pollutant removal in wastewater from ships was studied and optimal operating conditions were determined. The results show that the combination of treatment technologies has improved the reaction rate by 25% in the thermodynamic and kinetic models of pollutant removal, moreover, pollutant removal efficiency has been improved by nearly 20% compared with single technologies.

KEYWORDS

Wastewater from ships, water treatment, combined technology, activated carbon, photocatalyst, ozonation.

1. INTRODUCTION

Economic globalization is an inevitable trend, covering most fields and The global shipping industry is making breakthrough developments in the volume of goods transported. Ocean transportation carries more than 80% of the world's cargo and transfers about 3-5 billion tons of ballast water globally each year (Chu et al., 2020). A similar volume can also be transferred domestically between countries and regions each year. Ballast water is essential for the safe and efficient operation of modern shipping, providing balance and stability to unladen ships (Lakshmi et al., 2021). However, it can also generate a serious ecological, economic, and health threat to marine life. Moreover, the risks of marine environmental pollution due to illegal discharge of wastewater, oil spill accidents are becoming a serious environmental problem (Al-Tawaha et al., 2018; Nizetic et al., 2022). Wastewater from ships includes black water and gray water. Blackwater means sewage from ships and medical sinks. Gray water refers to wastewater from laundry, showroom, washbasins, showers, and sinks, which can be discharged directly into most marine environments without further treatment according to current regulations, which is Annex IV of the International Convention for the Prevention of Pollution from Ships (MARPOL) in force since 2003 (Han et al., 2019; Pham and Hoang, 2020). Operations to wash fuel tanks of the ships containing petroleum products and bilge water have been considered as the main source of marine pollution from ships (Hoang et al., 2018). The discharge of oily wastewater into the marine environment is a matter of scientific and technical concern. This problem is related to the harmful toxic effects that these substances can have on the environment and human health. Large amounts of oily wastewater can reduce the quantity and diversity of marine ecosystems (Cicek, 2019). Toxic compounds in oily wastewater can disrupt the structure of aquatic communities and food chains. Furthermore, toxic compounds also have various adverse effects on the surrounding environment, such as air pollution due to the evaporation of oil and hydrocarbon components into the atmosphere (Nizetic et al., 2022). In addition, they can affect groundwater, seawater, or drinking water. On the other hand, a large amount of oil spilled on the sea surface can cause fire and affect transportation safety. For these reasons, the International Maritime Organization (IMO) has issued the MARPOL convention 73/78 wherein wastewater from ships is limited to maximum oil content of 15 ppm (Bergesen et al., 2018; Pham et al., 2020). To conserve marine resources, current legislation has classified greasy wastewater generated from shipping operations as special and hazardous waste, thus imposing rules strict to handle it (Nizetic and Rowinski, 2021).

There are many proposed techniques for treating pollutants in ship waste streams based on physical, chemical, and biological methods. The vessel is equipped with an oil-water separator that helps to separate a large amount of oil (Hoang et al., 2021; Chau et al., 2021). However, their effectiveness results are often incomplete because when sediments are present, they can release more than 15 ppm of oil into the sea. Furthermore, the time-consuming separation process can promote the irreversible dissolution of organic compounds and the formation of emulsions that can occur in the presence of detergents (Bui and Pham, 2018; Chong et al., 2021). Among the physical methods, adsorption on

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activated carbon has been proposed (Kyzas et al., 2019; Vo et al., 2021). Activated carbon can produce high-quality wastewater, with oil content less than 10 mg/L and very low values of dissolved organic matter. Physical and chemical activation are the two main methods for the creation of activated carbon (de Franco et al., 2018). The physical activation of carbon is usually accomplished through a two-step process. In the first step, the precursor of carbon is pyrolysis at a relatively low temperature in the absence of air. As a result, materials with moderate surface area showed almost no pollutant absorbing activity (Shimizu et al., 2018). The second step involves activating the resulting coal in an oxidizing environment to significantly increase its surface area so that porosity develops which enhances pollutant removal from the environment. However, this physical treatment method is not capable of mineralizing the contaminants but only separates causing serious problems for their disposal (Wang et al., 2021). Furthermore, carbon needs to be replaced or regenerated after saturation. Recently, new attention has been drawn to the development of efficient processes for the removal of contaminants present in aqueous solutions with high salt content (Yetri et al., 2020; Menya et al., 2018).

The use of advanced oxidation processes can support well in the removal of petroleum mineralization and pollutants present in oily wastewater. These processes are based on the generation of hydroxyl radicals which are strong oxidants to mineralize organic pollutants. Heterogeneous photocatalysts are considered an emerging candidate based on the principle of non-selective oxidation that has been successfully used to oxidize many organic pollutants present in water systems (Wang et al., 2019; Pham et al., 2021). Among advanced oxidation processes, photocatalysis is a proven method that can be used in various environmental cleaning applications. Among the types of photocatalysts, TiO_2 is the most commonly used semiconductor photocatalyst for the conversion of organic pollutants into harmless ones (Reddy et al., 2020; Chitsomboon and Koonsrisook, 2021). Various materials have been explored as TiO_2 promoters for the photodegradation of contaminants in polluted water. Immobilization can be performed on a non-transparent substrate (such as activated carbon). In conventional water treatment, activated carbon (AC) is used to remove dyes by adsorption (Yang et al., 2019; Bui et al., 2021). Its characteristic is that it is extremely porous and has a very large surface area. These properties of activated carbon have attracted much research as a potential aid in photochemical processes. Activated carbon is also studied as a carrier for heterogeneous catalysis or in combination with TiO_2 for photocatalysis or as a composite material (Hao et al., 2018).

Ozone has been used as a disinfectant in drinking water since the late 1800s. Ozone is an excellent biocide and is unstable in water. Ozone (O_3) is one of the strongest oxidants and it is used to aid in the mineralization of organic pollutants from wastewater (Wu et al., 2018). Ozone is also being used more and more in wastewater treatment as an oxidizing and disinfecting agent. O_3 in combination with semiconductor catalyst and near-UV radiation has been successfully used to effectively treat organically polluted waters (Malvestiti et al., 2019). However, ozonation of seawater oxidizes bromide, which is a weaker, but more stable disinfectant (Bein et al., 2021). The half-life of ozone in water with a high concentration of Br ions was demonstrated to be 5s with the first step being a quantitative conversion to bromine. Because ozone decomposition is essentially quantitative at high Br-ion concentrations, the second important difference between freshwater and seawater is the absence of hydroxyl radical formation in seawater (Kharel et al., 2020). Therefore, the ozone treatment of marine ballast water involves an unstable primary oxidant, which is ozone, and a more stable secondary oxidant, which is bromine. Photogenerated electrons under UV irradiation can reduce BrO_3- ion to Br- ion, at the same time, the reverse reaction is hindered (Suryawan et al., 2019).

Each treatment method has both advantages and disadvantages, using a single method often results in low pollutant removal efficiency. Therefore, in this work, we propose an integrated treatment system, established by combining ozonation and photocatalysis with adsorption on activated carbon, to ensure the removal of at the same time and decompose organic impurities in polluted wastewater from ships. The complexity of bilge water has been simplified by using synthetic seawater in the presence of heavy metal ions. The advantage of combining different technologies is that they can overcome the disadvantages of separate usage methods, increase efficiency, and can reduce operating costs. The synergistic effect of the different methods enhances the oxidation rate of organic compounds during integration, so its use can yield sustainable results.

2. Materials and Methods

2.1 Materials

The experiments were carried out using a solution to simulate the wastewater of cargo ships transporting international routes from Vietnam to Japan. The raw wastewater samples used in the experiments were raw wastewater generated from the transport fleet. The simulated wastewater was prepared based on results obtained from the chemical characterization of the raw wastewater samples from different ships (WS1-WS6). The typical pollutant concentrations of the simulated wastewater samples are described in Table 1. In this study, we only report the analytical results of 6 wastewater samples. The results showed moderate variation for both inorganic and organic contaminant concentrations, mainly depending on the pretreatment processes used. The determined Cl ion content was similar for all samples and corresponds to typical concentrations in seawater (approximately 0.55 M). In this study, 4-nitrophenol (4-NP) was selected as the sample compound. It is one of 114 organic pollutants listed by the EPA in the United States and its maximum allowable concentration is 20 ppb. The photochemistry of 4-NP in the simulated wastewater sample was investigated as a function of various experimental parameters.

Table 1: Analysis of the composition of wastewater samples from ships

Samples	WS1	WS2	WS3	WS4	WS5	WS6
Pb^{2+}	0.008	0.058	0.076	0.095	0.052	0.065
Al^{3+}	0.065	0.785	0.086	0.732	0.062	0.685
Cd^{2+}	0.005	0.003	0.004	0.003	0.002	0.003
Cu^{2+}	0.145	0.058	0.125	0.155	0.045	0.145
Cr^{3+}	0.006	0.008	0.007	0.012	0.019	0.018
Co^{2+}	0.004	0.006	0.007	0.006	0.006	0.007
Mn^{2+}	0.065	0.055	0.062	0.035	0.032	0.028
Ni^{2+}	0.013	0.007	0.015	0.122	0.121	0.115
Fe^{3+}	0.036	0.068	0.158	1.025	2.555	0.168
Zn^{2+}	0.028	0.045	0.135	1.125	1.955	1.362
TOC	75.65	86.25	85.15	100.35	79.45	78.65

The Perkin-Elmer Optima 2100 ICP-OES analyzer in combination with the AS-90 autosampler is used to detect metal ions in samples. The wastewater concentration was set at 15 mg/ L. Nitrate salts are used as precursors of metal ions. The pH value of wastewater samples ranges from 4.5 to 5.5 depending on the concentration of the water sample. The influence of these parameters on metal concentrations will not be covered in this work. Metal ions strongly influence the kinetic model performed in this study, so their presence in the simulated wastewater is necessary. Notably, the number of heavy metal ions and organic substrates (called TOC values) used in the simulated wastewater was much higher than that obtained in the samples collected from the vessel. This selection is reasonable considering the possible extreme conditions and the high variability of contaminated wastewater from ships, therefore using an actual sample can often give fewer representative results than a simulated solution.

2.2 Methods

Batch experiments were performed in the presence of variable granular activated carbon (from 0.01 g to 0.15 g) in 25 mL of simulated wastewater containing 600 mg/L of 4-NP. The adsorption process reached adsorption equilibrium, adsorption experiments were carried out at 25°C, by stirring the solutions for 18 hours. Kinetic investigations were performed using a glass column containing a variable amount of absorbent material, between 1 and 15 g. Wastewater flow is set from 3 to 12 mL/ min. The established 4-NP flow varies from 250 to 1000 mg/L. All kinetic and equilibria investigations were supported by the intermediates in the presence of formic acid and acetic acid in photocatalytic oxidation or ozonation.

The photocatalyst used in this study is TiO_2 Aerooxide P25. The mixing molar ratio of Triethanolamine in 0.5 M tetraisopropoxide (TTIP) solution was 4. H_2O was added to the mixture after 2 hours of stirring to ensure an H_2O/TTIP molar ratio of 2. After stirring for 30 min, to avoid agglomeration, 20 grams of TiO_2 P25 was added slowly to 600 mL of the mixing solution. Then, the final TiO_2 suspension obtained was sonicated and stirred continuously for 24 hours in a closed tank. To increase surface roughness, Pyrex particles were kept for 24 hours in contact with an 8% hydrofluoric acid aqueous solution. Then, the granules were washed several times with distilled water, dried in an oven at 65°C, cooled at 25°C, and immersed in a TiO_2 suspension for about 50 min. The supernatant was separated by a funnel, then the granules were dried at 155 °C for 160 min

and calcined at 650°C for 3 hours to obtain the first coating. This procedure was repeated six times to obtain the required stable thickness of TiO_2. All photochemical experiments were performed in a continuous annular Pyrex photoreactor. The mass of TiO_2 added to the reactor was 0.15 grams. All photochemical experiments were performed at a temperature of 25 °C and a pressure of 1 atm. Before starting irradiation, aerate oxygen or air for about 30 minutes to form a saturated solution. Tests were performed with different concentrations of 4-NP (50–200 ppm) in demineralized water.

O_3 is produced by supplying pure oxygen and He. The concentration of ozone in the gas phase is determined by iodine titration and ranges from 0.095 to 1.65 ppm respectively. The corresponding ozone concentrations in the liquid phase were calculated according to Henry's law and ranged between 0.084 and 0.35 ppm, respectively (Staudinger and Roberts, 1996). The measured concentrations of O_2 in the reaction solution are very close to the equilibrium concentrations and they are almost unchanged in the presence of ozone. Operations were performed starting at 200 ppm 4-NP in synthetic wastewater at 25°C and 1 atm pressure.

A study was performed by combining the previously described process with activated carbon adsorption. The adsorption rate experiments were performed by circulating the reaction solution through an adsorption column containing different amounts of granular activated carbon (ranging from 0.4 to 1.6 g). Then, a series of experiments varied the amount of granular activated carbon to determine the enhancement factor for the entire integration. Figure 1 shows a schematic diagram of photocatalytic combined activated carbon ozonation.

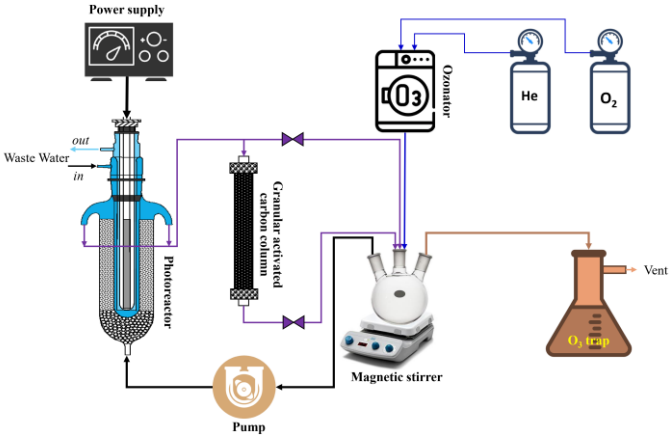

Figure 1: Wastewater treatment system combines three advanced processes

3. Results and Discussion

3.1. Wastewater treatment with granular activated carbon

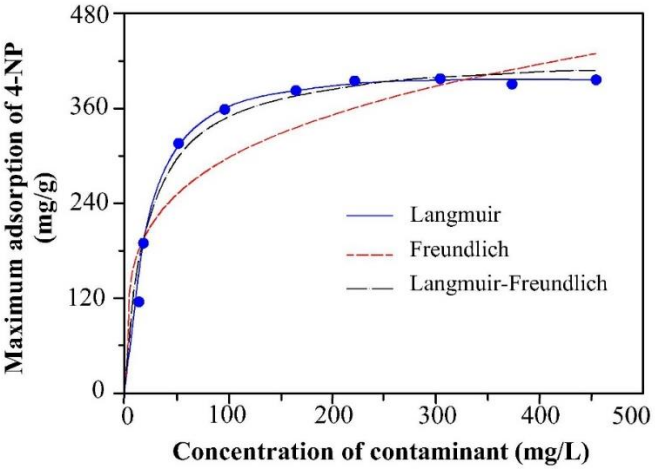

Figure 2: Evolution of the maximum adsorption of 4-NP on granular activated carbon

4-NP adsorption assays using granular activated carbon were used to evaluate the thermodynamic and kinetic parameters of the adsorption process. The selection of data to optimize the parameters of the oxidation optimization process in the integrated wastewater treatment system was carried out according to the evolution of the maximum adsorption of 4-NP on granular activated carbon. The set of such data is depicted in Figure 2, the experimental data are evaluated in the correlation of Langmuir, Freundlich, and Langmuir–Freundlich equations.

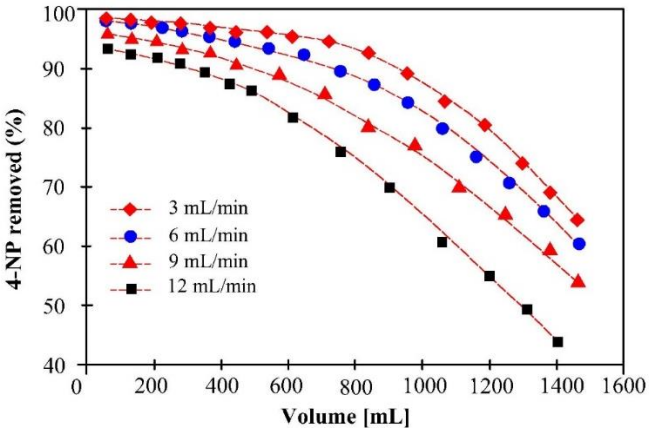

Figure 3: Correlation between 4-NP adsorption efficiency on activated carbon with different wastewater rate flow.

The optimization process used regression diagnostic techniques for the data of each trial. Model reliability is ensured through residual analysis for each regression. The graph depicts the observed residuals of the model of the Freundlich isotherm equation as the highest. It was revealed that the maximum absorption increased most strongly as the concentration of pollutants increased. This is true for materials with heterogeneous surfaces with low absorbent concentrations. The linear curves obtained from the kinetic model of the Langmuir-Freundlich equation and the Langmuir model are more suitable for the experimental data. Langmuir model shows the best correlation with experimental data. That means it is suitable and optimal to characterize the relationship of 4-NP adsorption capacity on granular activated carbon under the operating conditions established for the integrated system. Kinetic studies allow the establishment of optimal test conditions to improve the removal efficiency of pollutants in wastewater from ships. Evaluation of the ability to remove pollutants can be related to parameters such as wastewater flow, pollutant concentration, and active carbon content in the filter device. The data obtained from the tests show quantitative values for the saturation of activated carbon as well as the pollutant filtration efficiency of activated carbon adsorption. Figure 3 shows the effect of wastewater flow on the adsorption capacity of activated carbon by immobilizing the amount of granular activated carbon and the initial concentration of the pollutant. When the flow rate is higher than 9 mL/min, the removal efficiency of pollutants can be reduced because the contact time between activated carbon and wastewater is not large enough to ensure complete adsorption.

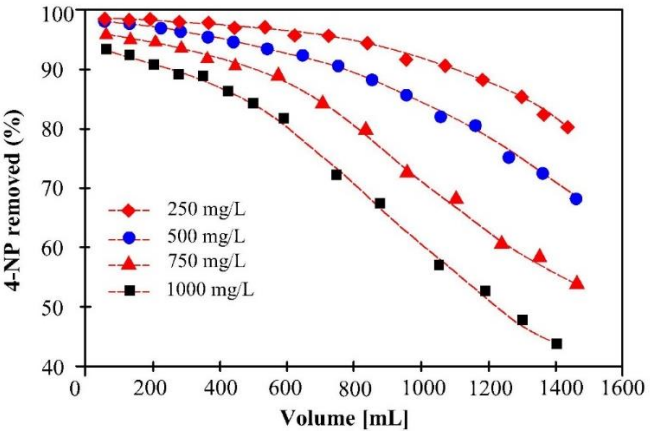

Figure 4: Correlation between 4-NP adsorption efficiency by volume of wastewater with different initial wastewater concentrations

Figure 3 also revealed that the pollutant removal efficiency decreased significantly when the wastewater volume was greater than 800 mL with wastewater flow rates at 9 mL/min and 12 mL/min. With lower wastewater flow rates (at 3 mL/min and 6 mL/min), the volume of treated wastewater is doubled. The trend of change in pollutant removal efficiency at 3 mL/min and 6 mL/min was quite similar. Therefore, a wastewater flow rate at 6 mL/min was chosen for the following experiments as it

represents a suitable balance between removal efficiency and the time required to achieve it.

Figure 4 showed that the initial pollutant concentration affected the 4-NP absorption efficiency on activated carbon. The integrated system using activated carbon can completely filter pollutants with wastewater concentrations of 250 mg/L and 500 mg/L. The removal efficiency was above 90% averaged for the initial wastewater concentration of 250 mg/L. However, when the initial wastewater concentration increased, the pollutant removal efficiency decreased sharply, especially when the wastewater volume exceeded 800 mL. Notably, although the adsorption curve does not show rejection values close to zero, which can be interpreted as a fully saturated material, the efficiency results are too low for practical applications.

3.2 Wastewater treatment with heterogeneous photocatalysis

The efficiency of pollutant removal in wastewater by the heterogeneous photocatalyst system is described by the concentration of 4-NP absorbed by the photocatalyst according to the contact time, with wastewater concentrations set from 50 mg/L to 200 mg/L.

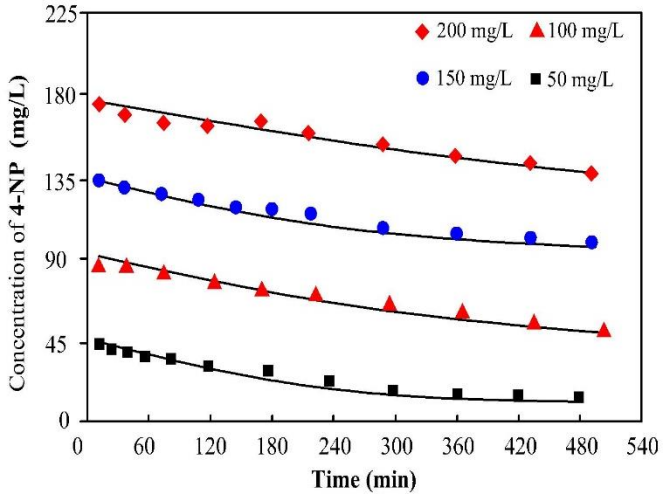

Figure 5: Change in 4-NP concentration in demineralized wastewater over time of tested heterogeneous photocatalyst exposure

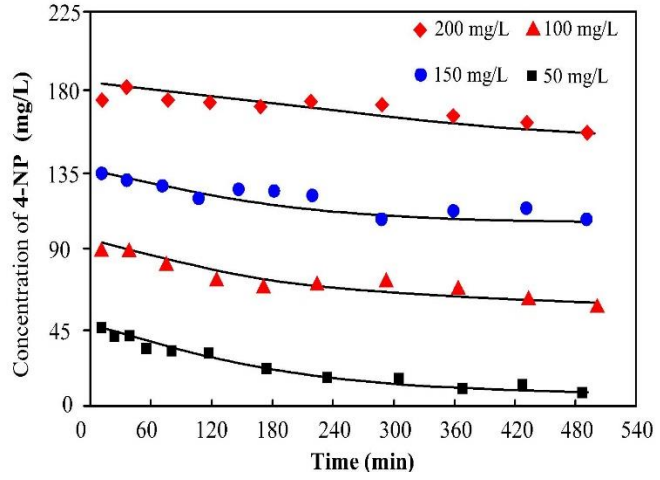

Figure 6: Change of 4-NP concentration in simulated wastewater over time of tested heterogeneous photocatalyst exposure

In Figures 5 and 6, the results show that as the contact time of wastewater with photocatalysts increases, the absorption concentration of 4-NP decreases. It can be observed that the conversions in the case of demineralized wastewater have significantly reduced concentrations of pollutants. The ionic strength of the solution reduces the solubility of oxygen. Moreover, chloride ions present in the solution act as OH radical scavengers, so the photocatalytic reaction rate is very low. The mechanism of the photocatalyst reaction has revealed that when the light of suitable energy shines on the TiO_2 surface, the solid occurs excitation and generates electron-hole pairs on the surface. These charges can be trapped by substances on the TiO_2 surface giving rise to highly oxidizing substances such as hydroxyl radicals, thereby initiating oxidation reactions.

3.3 Wastewater treatment with homogenized ozonation

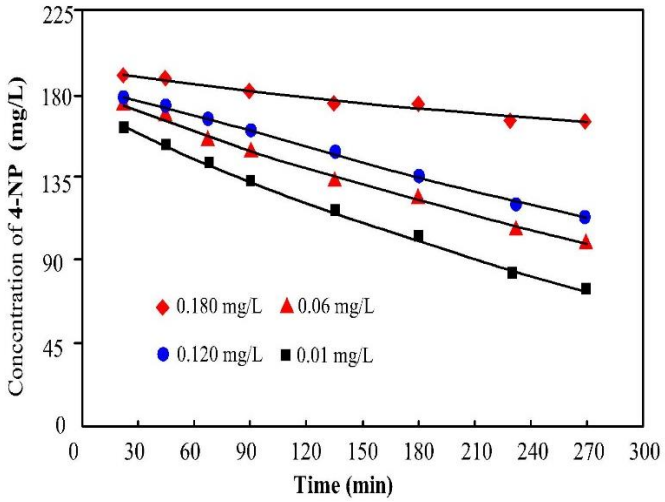

Figure 7: Change in 4-NP concentration of laboratory wastewater over time in ozone with different concentrations

Initial experimental steps indicated that 4-NP concentrations remained constant during O2 bubbling in solution and/or during UV-A irradiation. The addition of ozone in the reaction solution resulted in a decrease in 4-NP concentration and this reduction was not affected by UV-A irradiation. Figure 7 reports the values of 4-NP concentrations versus reaction times for runs performed in the presence of different concentrations of ozone in the liquid phase. It can be observed that the reaction rate increases with the increasing concentration of dissolved ozone. Ozone decomposes slowly in the wastewater solution through multi-step reactions leading to the formation of oxidizing radicals. In the presence of organic or inorganic compounds, ozone reacts through two pathways. The first is a direct selective reaction with specific functional groups and the second is a reaction with free radicals generated from ozone decomposition.

3.4 Wastewater treatment by combining three advanced processes

The limitations of each technology are overcome by the solution of coupling activated carbon columns with photocatalytic ozonation allowing the best correlation between the three processes. Indeed, the lifetime of activated carbon can be enhanced in the presence of ozone, which is capable of in situ oxidizing resurfacing adsorbed molecules. Furthermore, small pollutant molecules are not normally captured by activated carbon, so combining with some advanced oxidation processes can avoid the release of these compounds. On the other hand, the presence of activated carbon allows the treatment of higher wastewater output than for single photocatalytic ozonation. Furthermore, the ozonation rate is reported to increase on the surface of activated carbon. The optimization problem was approached by varying the amount of activated carbon to highlight its influence on the coalescence process.

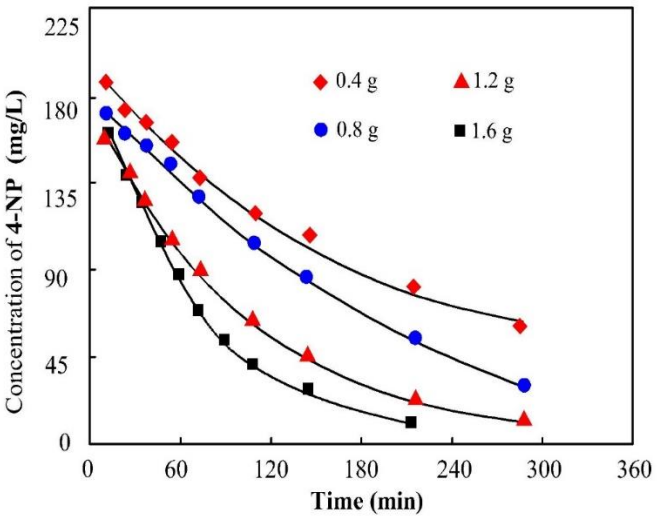

Figure 8: Change in 4-NP concentration of wastewater simulated over time in a system combining ozone and activated carbon with variable activated carbon volume

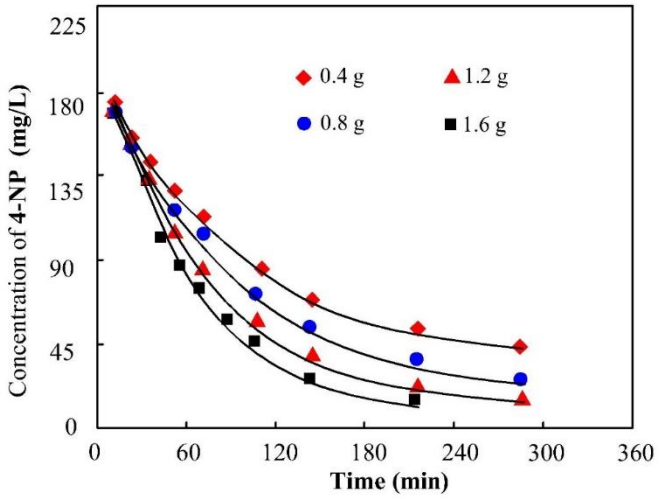

Figure 9: Change of 4-NP concentration of wastewater simulated overtime performed in a system combining TiO2 and activated carbon with variable activated carbon volume

Figure 8 shows a series of tests performed by varying the amount of activated carbon in the absence of photocatalytic ozone. It can be seen that the data described in terms of exponential curves have a good correlation. Tests were performed to determine the synergistic effect when adsorption by activated carbon and photocatalytic ozone were combined. Figure 9 depicted the experiments performed under the determined optimal conditions for photocatalytic ozonation with different amounts of activated carbon in the column. It can be seen that the exponential curves are in good agreement with the experimental data. In this way, it is possible to quantify the effect of the amount of activated carbon on the combined process using the previously described procedure.

4. Conclusion

Disposal of marine waste is a challenging issue for marine environmental management strategies because single decontamination processes can present several problems. The photocatalysis mechanism is strongly inhibited by the presence of salts in seawater, which adversely affects the oxygen solubility as well as competes with organic substrates for adsorption on the surfactant sites. On the other hand, the formation of BrO_3 ions during the oxidation of polluted seawater ozone has a high potential for the formation of carcinogens. Therefore, it is essential to combine the above two advanced oxidation processes to improve the reaction rate and reduce toxic by-products. Moreover, for polluted water containing many organic components, the implementation of granular activated carbon adsorption improved the ability to filter organic matter as well as stimulated ozonation to occur on the surface of the water. In contrast, granular activated carbon surfaces can be partially recovered by partial ozonation of the adsorbed pollutants. That was able to prolong the life of the absorbent material. This study has focused on assessing the ability to remove pollutants in the wastewaters from ships by the combination of advanced treatment methods mentioned above. In particular, to enhance the synergistic effect of granular activated carbon, photocatalysis, and ozonation in wastewater treatment, optimal conditions were established to select the best thermodynamic and kinetic model. Synergistic effects between photocatalysis and ozonation were observed even under the considered high salinity conditions. Experimental data have shown that the combination of ozonation and photocatalysis results in a 25% improvement in the reaction rate compared to the total rate in comparison with the single technologies. The combination of photocatalytic ozonation with adsorption on granular activated carbon not only brings about the advantages reported above but also leads to an increase in the treatment efficiency of the whole system. The synergistic effects increased the reaction rate by 18% compared to the total speed of the single technologies mentioned above. This work has allowed us to effectively rationalize the mutual necessity of different technologies applied to wastewater from marine vessels.

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