ORIGINAL PAPER

Kinetic study of the photocatalytic degradation of the acid blue 113 dye in aqueous solutions using zinc oxide nanoparticles immobilized on synthetic activated carbon

Abdollah Dargahi[1,2], Mohammad Reza Samarghandi[3,✉], Yaser Vaziri[1,2], Ghobad Ahmadidoust[1], Esmail Ghahramani[2], Ali Akbar Shekarchi[4]

1. Student Research Committee, Hamadan University of Medical Sciences, Hamadan, Iran
2. Department of Environmental Health Engineering, School of Health, Hamadan University of Medical Sciences, Hamadan, Iran
3. Research Center for Health Sciences and Dep. Environmental Engineering School of Public Health, Hamadan University of Medical Sciences, Hamadan, Iran
4. Department of Public Health, Khalkhal University of Medical Sciences, Khalkhal, Iran

Date of submission: 22 Dec 2018, ***Date of acceptance:*** 16 Feb 2018

ABSTRACT

Approximately 10-20% of the total dyes in the world is consumed in the textile industry. The present study aimed to investigate the photocatalytic activity of zinc oxide nanoparticles (ZnO) immobilized on synthetic activated carbon in the removal of the acid blue 113 dye from aqueous solutions. This experimental study was conducted in a photo-reactor with the useful volume of one liter. The effects of pH (3, 7, and 9), zinc oxide nanoparticle concentrations (0.1-0.4 mmol/l), concentration of activated carbon modified by zinc oxide nanoparticles (20, 40, 60, 80, and 100 mg/l), and the initial concentration of the dye (20, 40, 60, 80, 100, and 200 mg/l) were assessed. In addition, the kinetics of the reaction were investigated. The results indicated that the optimal conditions for the process were the pH of 3, activated carbon modified by zinc oxide nanoparticle concentration of 100 mg/l, ratio of 0.4 millimole of zinc oxide per gram of activated carbon, and acid blue 113 dye concentration of 100 mg/l, which resulted in the maximum efficacy of 96%. Moreover, removal efficiency using zinc oxide was greater in all the stages compared to removal efficiency using activated carbon. The kinetic rate was also determined, demonstrating that the process followed the first-order kinetics. In addition, the findings indicated that the process had outstanding efficiency in the removal of the acid blue 113 dye. The photocatalysis of nanoparticle oxidation on synthetic activated carbon could be used effectively as an advanced oxidation reaction to remove dyes.

Keywords: Photocatalytic Process, Zinc Oxide, Synthetic Active Carbon, Acid Blue 113, Aqueous Solutions

Introduction

Today, population growth, development of various industries, increased drought, and decreased available water resources for humans for domestic and agricultural uses are the major risk factors for the pollution of water resources and unprecedented shortages.[1,2] The continuation of this process has urged the treatment and recycling of water resources. Among various industries, textile industries have expanded across the world, especially in Iran. The dying units in the textile industry are considered to be the major causes of water resource and environmental pollution due to the consumption of thousands of dye chemicals.[3]

Water consumption is also extremely high in textile industries, which leads to the production of large volumes of wastewater containing toxic and refractory organic compounds, which are potentially hazardous to the environment.[4, 5] The textile industry is one of the largest dye consumers in the world, so that approximately 10-20% of the total consumed

✉*Mohammad Reza Samarghandi*
samarghandi@umsha.ac.ir

Citation: Dargahi A, Samarghandi MR, Vaziri Y, Ahmadidoust Gh, Ghahramani E, Shekarchi AK. Kinetic study of the photocatalytic degradation of the acid blue 113 dye in aqueous solutions using zinc oxide nanoparticles immobilized on synthetic activated carbon. J Adv Environ Health Res 2019; 7(2): 75-85

dyes in the world is consumed in the textile industry.

Discharging colorful sewage into the environment, especially water resources, disrupts the aesthetic and ecological aspects, leading to the phenomenon of eutrophication and algal bloom in surface waters; this is one of the most significant issues associated with the discharge of dyes into the environment.[6, 7] The presence of organic dyes in the wastewater of textile industries and lack of proper treatment systems leads to the pollution of water resources and poor quality of surface and underground waters.

The acid blue 113 dye is a synthetic dye, which is resistant to sunlight and various treatment processes.[8] Due to its cost-effectiveness, the use of this dye has increased in the textile industry. The decomposition of this dye in the environment is toxic and mutagenic.[8] The conventional methods used for the treatment of textile wastewater include biological, physicochemical, and electrochemical treatment processes and advanced oxidation.

Advanced oxidation processes are based on the production of strong oxidizing radicals (e.g., hydroxyl radical, sulfate radical, superoxide radical, and hydroperoxyl radical), which have a high tendency to degrade pollutants.[9, 10] In photocatalytic degradation, pollutants are completely decomposed and destroyed using an optical source (UV light).[11-13]

In heterogeneous catalytic processes, the reaction materials are not in a similar phase and cannot simply dissolve in the reaction medium. In order to prevent the reduction of the reaction efficiency caused by decreased active surface, a substrate is applied as a catalyst. In the present study, porous active carbon derived from mango seed was used for this purpose.[14, 15] Figure 1 shows a number of advanced treatment techniques, as well as the limitations of each method.[10]

In a study by Shu *et al.*, the effect of ultraviolet radiation on the activation of persulfate as an oxidizing agent was assessed in the removal of the acid blue 113 dye. According to the findings, the dye removal efficacy was higher with the simultaneous use of persulfate and ultraviolet radiation compared to the independent use of persulfate and ultraviolet radiation.[16] In addition, maximum dye degradation was achieved at the initial persulfate concentration of 6.6 millimoles. Moreover, increasing the initial concentration of the acid blue 113 dye was reported to decrease dye degradation.

According to the findings of Shu H-Y *et al.*, initial pH had no significant effect on the removal efficiency of the dye.[16] Muthirulan *et al.* studied the photocatalytic activity of zinc oxide (ZnO) photocatalyst supported by porous activated carbon in the degradation and mineralization of alizarin cyanine green dye from aqueous solutions, observing that changed in the pH of the environment from acidic to neutral and alkali conditions diminished the efficiency of the process, so that the maximum efficiency was achieved in acidic conditions, and the minimum efficiency was denoted in alkali conditions.[17] Moreover, after increasing the initial concentration of the dye and due to the reduced ultraviolet penetration and active surface of the photocatalyst, the highest efficiency was achieved at low initial concentrations.[17]

Given the importance of the removal of dyes from the colorful wastewaters discharged by industries, such as the textile industry, the present study was the first to assess the potential of zinc oxide nanoparticles immobilized on synthetic activated carbon derived from mango seeds in dye removal from aqueous solutions. In addition, we aimed to investigate the photocatalytic activity of zinc oxide nanoparticles immobilized on synthetic activated carbon derived from mango seeds in the removal of the acid blue 113 dye from aqueous solutions.

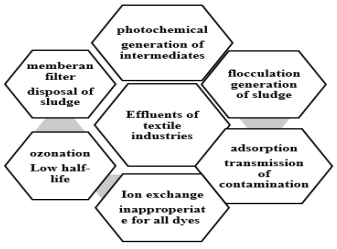


Fig. 1. Advanced treatment methods and their limitations

Materials and Methods

Experimental materials

In this study, the acid blue 113 dye (purity: 99.9%) was purchased from Sigma-Aldrich Company, United States. The schematic view and features of the reactor used in the experiment are shown in Figure 2 and Table 1, respectively. The studied samples included synthetic effluents, which were made in a laboratory with various concentrations of the acid blue 113 dye. The properties of the acid blue 113 dye are presented in Table 2.

Measurement of the acid blue 113 dye was carried out at the experimental stage using a spectrophotometer apparatus at 568 nm. The other chemicals used in the experiments included sodium hydroxide, sulfuric acid, and zinc oxide nanoparticles, which were obtained from Sigma-Aldrich Company. The number of the samples was calculated based on the one-factor-at-a-time method, and 32 samples were prepared. The results were analyzed using the Minitab software version 17 and Excel software 2016. The removal efficiency of the acid blue 113 dye was determined using the following equation:[18, 19]

$$\text{Efficiency}=\frac{C1-C2}{C1}$$

where C1 is the initial concentration of the acid blue 113 dye (mg/l), and C2 represents the final concentration of the acid blue 113 dye (mg/l).

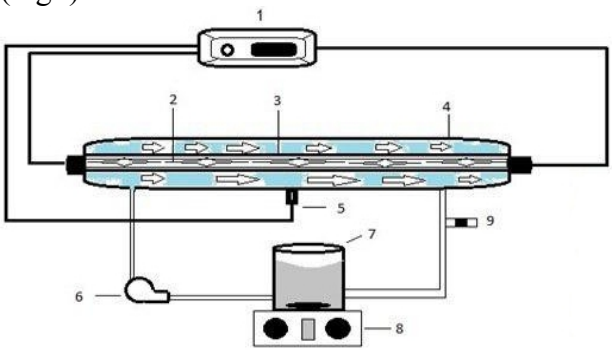
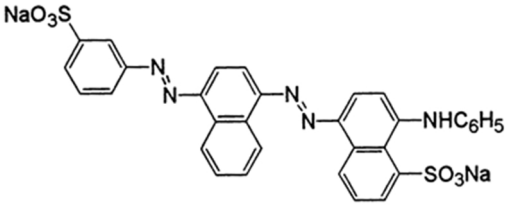
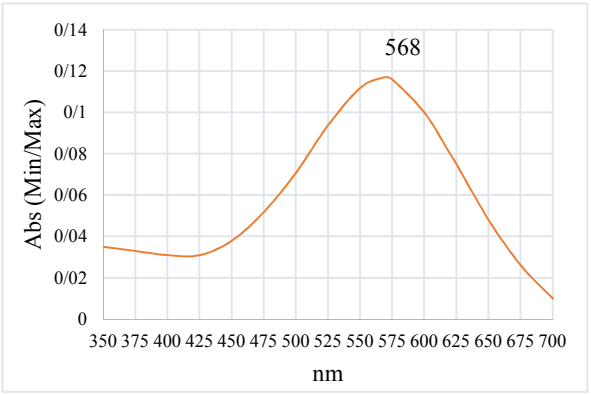


Fig. 2. Schematic view of reactor; 1) transformator; 2) low pressure Hg UV lamp; 3) quartz jacket; 4) stainless steel jacket; 5) photocell; 6) pump; 7) sample vessel; 8) shaker; 9) sampling tube

Table 1. Features of reactor

Type of reactor	Photo-chemical
Shape of the reactor	Cylindrical
Chamber material	Stainless steel
Useful volume	1 L
Lamp power	55W

Table 2. Properties of acid blue 113 dye

Type of dye	Acidic, soluble, azo dye
Molecular structure	
Chemical formula	$C_{32}H_{21}N_5Na_2O_6S_2$
Molecular weight	681.649 g/mol
Wavelength scan	

Experimental methods

Production and modification of activated carbon with zinc oxide

At this stage, 50 grams of dried mango seeds were mixed with a specific volume of phosphoric acid (concentration: 95%) with the mass ratio of 1:10. The resulting mixture was placed in an electric furnace at the temperature of 700 ºC for one hour. Afterwards, the furnace was turned off until its temperature reached an ambient temperature. The produced carbon was washed with distilled water until the pH exceeded 6.5.

In order to dry the carbon, it was placed in an oven again at the temperature of 120 ºC. After drying, the carbon was crushed using a porcelain pestle and filtered through the mesh of 30, 40, and 50 sieves. Modification of the activated carbon was performed using the mechanical-hybrid method at the mass ratio of the zinc oxide nanoparticles to the activated carbon with the concentration of 0.1, 0.2, 0.3, and 0.4 mmol/g. In addition, scanning electron microscopy (SEM) and X-ray diffraction (XRD) were performed on the activated carbon samples before and after modification by zinc oxide so as to ensure the modification of the activated carbon.

Determining the optimal pH

In order to determine the initial pH for the efficacy of the dye removal process, samples (volume: 1 l) with the initial pH of 3, 7, and 9 were prepared and introduced into the reactor. The adjustment of pH was carried out using 0.1 N NaOH and H_2SO_4. At this stage, the other parameters remained constant (initial dye concentration: 100 mg/l, modified photocatalyst concentration: 100 mg/l, reaction time: 30 minutes). Finally, the pH associated with the maximum removal efficiency was selected as the optimal pH.

Determining the modified photocatalyst concentration

After determining the optimal pH for maximum dye removal efficiency, the other parameters remained constant (initial dye concentration: 100 mg/l, modified photocatalyst concentration: 100 mg/l, reaction time: 30 minutes), and various concentrations of modified activated carbon (20, 40, 60, 80, and 100 mg /l) were added to the samples. The initial concentration associated with maximum removal efficiency was selected as the optimal concentration of the modified photocatalyst.

Determining the initial ratio of zinc oxide to activated carbon

After obtaining the optimal pH and modified photocatalyst concentration, the optimal concentration of zinc oxide was determined by altering the ratio of zinc oxide to activated carbon with the active carbon concentrations of 0.1, 0.2, 0.3, and 0.4 mmol/g in constant conditions (initial dye concentration: 100 mg/l, modified photocatalyst concentration: 100 mg/l, reaction time: 30 minutes).

Effects of the initial concentration of the acid blue 113 dye

At this stage, the optimal concentration of the acid blue 113 dye was investigated. In order to determine the optimum value, the initial dye concentrations of 20, 40, 60, 80, 100, and 200 mg/l were selected, and the optimal initial dye concentration was assessed with the other parameters remaining constant (pH=3, modified photocatalyst concentration: 100 mg/l, reaction time: 30 minutes). Finally, the concentration associated with the maximum removal efficiency was considered as the optimal concentration.

Effects of zinc oxide alone and zinc oxide immobilized on activated carbon

At this stage, the effects of zinc oxide alone and zinc oxide immobilized on activated carbon on the removal of the acid blue 113 dye were evaluated. To this end, in the optimal conditions of the experiments (pH=3, initial dye concentration: 100 mg/l, modified photocatalytic concentration: 100 mg/l, reaction time: 30 minutes), the zinc oxide nanoparticle concentrations of 0.1 and 0.4 mmol/g were used to investigate the dye removal efficiency with zinc oxide alone and compare the results with those obtained with the application of zinc oxide

immobilized on activated carbon.

Comparison of the overall efficiency for systems

Based on the results obtained in the previous steps and after determining the optimal values of each parameter involved in the experiments, the effects of the influential factors on the efficiency of the main process were assessed. In optimum conditions, the effects of ultraviolet radiation, zinc oxide in the presence of ultraviolet radiation, and zinc oxide immobilized on activated carbon in the presence of ultraviolet radiation were determined.

Determining the kinetics of the reaction

In chemical sciences, determining the reaction rate is considered essential to the process. Therefore, the kinetics of the reaction were evaluated in order to determine the function of the pollutant removal reaction, as well as to model and execute the process on an applied scale. At this stage and in optimum conditions as in the previous stages, the kinetic models were prepared, and the zero-, first-, and second0order kinetic models were applied. Finally, the optimal kinetic model for the process was determined based on the obtained correlation-coefficients.

Results and Discussion

Analysis of the produced activated carbon and modified by zinc oxide

Analysis of the synthetic and modified photocatalyst preparation methods is crucial to the experiments. At this stage, the laboratory analysis of the produced activated carbon from mango seeds modified with zinc oxide nanoparticles was carried out. The results of the specialized SEM test (Figure 3) indicated increased porosity on the surface of the synthesized activated carbon. Furthermore, the results showed that the zinc oxide nanoparticles were located on the surface and inside the porous synthesized active carbon particles.

XRD is another test performed in the AC-ZnO composite, which was conducted in this study in order to verify the accuracy of the positioning of zinc oxide (Figure 4). The results indicated the presence of the peaks at the temperatures of 31.6 ºC, 34.2 ºC, 36.2 ºC, and 47.4 ºC in zinc oxide.[17, 20, 21]

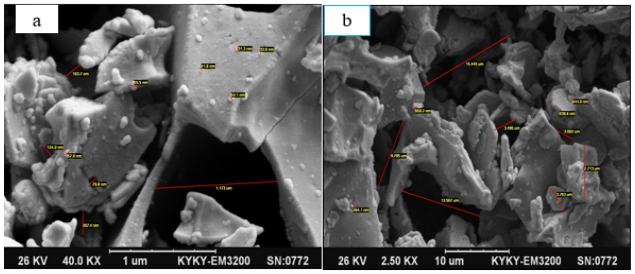


Fig. 3. SEM images of activated carbon; a) unmodified; b) modified with ZnO nanoparticles

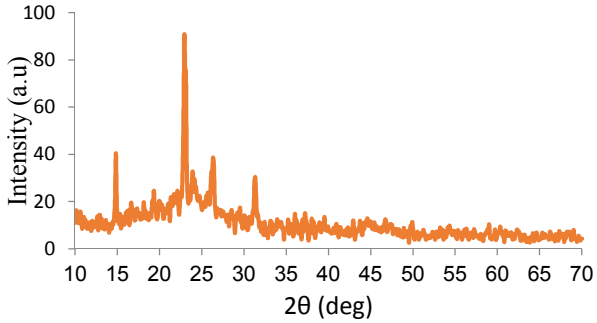


Fig. 4. XRD pattern of activated carbon modified with ZnO nanoparticles

Effect of pH

In general, pH is considered to be the most reliable parameter to determine the efficiency of chemical processes since pH directly affects process efficiency through influencing the surface charge. The obtained results of the present study in this regard are shown in Figure 5. Accordingly, the system was most efficient in acidic conditions, where the maximum removal efficiency was estimated at 96%, while in alkaline conditions, the minimum efficiency was estimated at 45%. In acidic conditions, the surface of photocatalytic zinc oxide had a positive charge, which significantly enhanced the removal efficiency of the acid blue 113 dye, thereby increasing the decomposition and removal efficiency.[3, 16]

In acidic and neutral conditions, various events were observed, and degradation efficiency decreased.[3, 16] On the other hand, one of the important features of each adsorbent is the point of zero charge (PZC), which was measured in the synthesized carbon and zinc oxide nanoparticle in this study. The obtained

values for activated carbon and nanoparticle oxidation were 6 and 6.25, respectively.[22] At the pH of lower than the PZC, the surface of the molecule was positively charged due to the presence of H^+ ions, thereby increasing the adsorption of the acid blue 113 dye. However, at higher pH values, the electrical charge on the surface of the zinc oxide nanoparticle was negative due to negative OH^- ions.[22]

In a study in this regard, Muthirulan *et al.* investigated the role of zinc oxide photocatalyst supported with porous activated carbon for the mineralization of alizarin cyanine green dye in aqueous solutions. According to the findings, changes in the pH of the environment from acidic to neutral and alkaline decreased the efficiency of the process, so that the maximum efficiency was achieved in acidic conditions, and the minimum removal efficiency was obtained in alkaline conditions.[17] In another research conducted by Shu HY *et al.*, the effect of ultraviolet radiation on the activation of persulfate as an oxidant in the removal of the acid blue 113 dye was investigated, and the results indicated the maximum acid blue 113 dye removal efficiency in acidic conditions at two and 10 minutes to be 90.8%.[16]

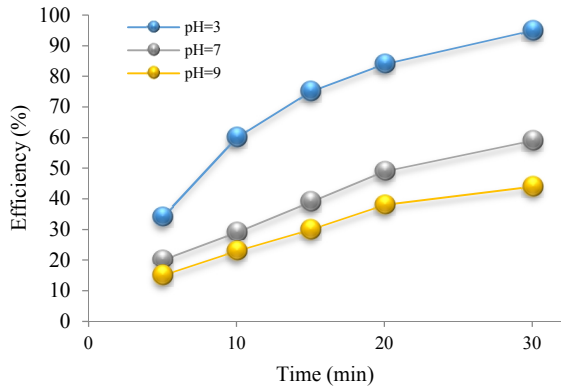


Fig. 5. Effect of initial pH changes on removal of acid blue 113 dye (initial dye concentration: 100 mg/l, modified photocatalyst concentration: 100 mg/l, reaction time: 30 minutes)

In another study, Arbabi *et al.* evaluated the removal of the acid blue dye using the UV/H_2O_2/Fe_3O_4 process through the optimization of the treatment process based on an experimental design. The obtained results indicated that the maximum removal efficiency of the acid blue 113 dye in optimum conditions (pH: 3.8, initial dye concentration: 62.3 mg/l, H_2O_2 concentration: 31.33 mmol/l, reaction time: 59.5 minutes, Fe_3O_4 concentration: 81.1 g/l) was 95.51%, which is consistent with the results of the present study.[23, 24]

Effect of changes in photocatalyst concentration

The findings of the current research showed a direct correlation between the initial concentration of the photocatalyst and removal efficiency of the acid blue 113 dye. Accordingly (Figure 6), with the minimum and maximum photocatalyst concentration of 20 and 100 mg/l, the minimum and maximum removal efficiency was estimated at 45% and 96%, respectively. Moreover, the increased initial concentration of the modified photocatalyst increased the active sites on the surface of synthesized activated carbon, thereby incrementing the number of the active sites and acid blue 113 dye adsorption, which in turn enhance the removal efficiency.[17, 25, 26] Our findings in this regard are in line with the study by Kadirova *et al.*, in which photocatalytic degradation and the adsorption of methylene blue from saline solutions were assessed using the activated carbon-coated iron ion.[25] In addition, Muthirulan *et al.* reported that increasing the modified catalyst led to higher process efficiency, and as the catalyst increased by 280 mg/l, the process efficiency increased by 95%. This is consistent with the results of the present study.[17]

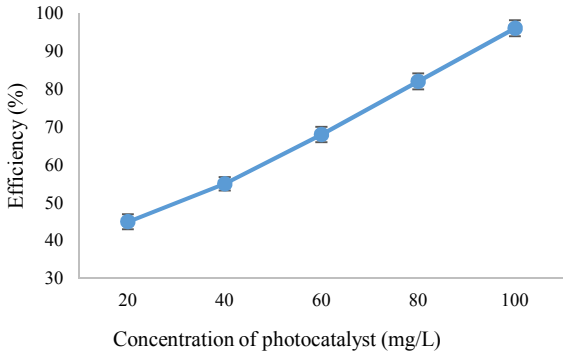


Fig. 6. Effects of photocatalytic concentration changes on removal of acid blue 113 dye (pH: 3, initial dye concentration: 100 mg/l, modified photocatalyst concentration: 100 mg/l, reaction time: 30 minutes)

Effect of the ratio of zinc oxide to activated carbon

With the increased concentration of nanoparticle oxidation, the system performance improved. As is depicted in Figure 7, the increased ratio of the initial concentration of zinc oxide to activated carbon enhanced the process efficiency, so that the process efficiency changed from 45% at the initial concentration of 0.1 mmol/g of oxidation to 96% at the initial concentration of 0.4 mmol/g of activated carbon. In this process, the nanoparticles absorbed the high-energy photons of the ultraviolet spectrum, releasing the active chemicals (e.g., free hydroxyl and peroxide radicals) that play a key role in the reaction and degradation of the pollutant.[17, 27]

According to a study by Samarghandi *et al.*, which aimed to assess the efficiency of the photocatalytic process of titanium dioxide in the removal the RB5 dye and cyanide, increased concentration of titanium dioxide photocatalyst enhanced the efficiency of the removal process, so that the maximum efficiency was obtained at the photocatalyst concentration of 1 gr/l.[28]

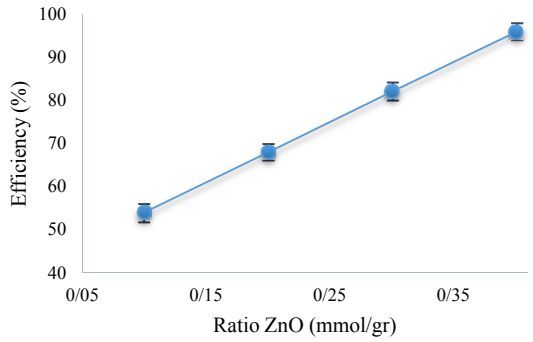


Fig. 7. Effect of changes in initial concentration of ZnO to activated carbon in removal of acid blue 113 dye (pH: 3, initial dye concentration: 100 mg/l, modified photocatalyst concentration: 100 mg/l, reaction time: 30 minutes)

Effect of the initial concentration of the acid blue 113 dye

The results of the experiments in this section are shown in Figure 8. Accordingly, the increased initial concentration of the acid blue 113 dye was associated with the higher number of the dye molecules that were adsorbed on the photocatalyst surface, avoiding the reactions between the dye molecules, produced photon holes, and hydroxyl radicals due to the lack of direct contact.[29]

According to the results of the present study, the increased initial concentration of the dye in the solution led to the higher consumption of the oxidizing agent and increased the duration of treatment. This is consistent with the results obtained by Rahmani *et al.* in the assessment of methylene blue removal via the nano-TiO_2/UV process.[30] In addition, the findings of Muthirulan *et al.* demonstrated that increasing the initial concentration of dye diminished the efficiency of the process, so that the removal efficiency was approximately 100% at the initial concentration of 10 mg/l, while in the same conditions and at the initial concentration of 80 mg/l, the removal efficiency reduced to 97%.[17]

In another research, Seid-mohammadi *et al.* investigated the degradation of the acid blue 113 dye using US/H_2O_2/Fe^{2+} and US/$S_2O_8^{2-}$/Fe^{2+}. According to their findings, the simultaneous use of both processes decreased the process efficiency with the initial concentration of the acid blue 113 dye, so that in optimum conditions, the maximum efficiency was obtained at the concentration of 25 mg/l, and the minimum efficiency was obtained at the highest concentration of the acid blue 113 dye (200 mg/l).[31]

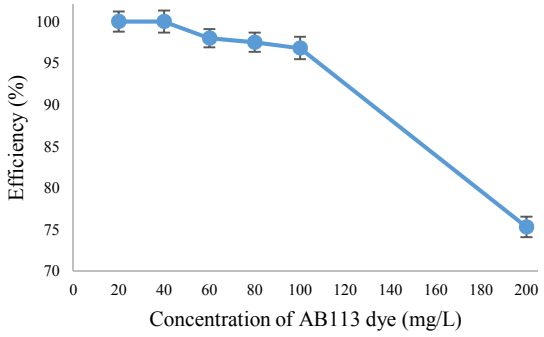


Fig. 8. Effect of initial acid blue 113 dye concentration on dye removal efficiency (pH: 3, modified photocatalyst concentration: 100 mg/l, reaction time: 30 minutes)

Effect of changes in zinc oxide alone and zinc oxide immobilized on activated carbon

At this stage, the changes in the process efficiency were assessed in two separate states

(i.e., in the presence of oxidation and in zinc oxide nanoparticles immobilized on activated carbon); the obtained results are depicted in Figure 9. According to the findings, zinc oxide alone had lower efficiency compared to the zinc oxide nanoparticles immobilized on activated carbon. Moreover, the process efficiency at the initial zinc oxide concentrations of 0.14-0.4 mmol/l was estimated at 58%, 44%, 30%, and 72%, while the efficiency using the zinc oxide nanoparticles immobilized on activated carbon was estimated at 82%, 68%, 54%, and 96%. Therefore, it could be concluded that the oxidation efficiency alone was lower at all the concentrations compared to zinc oxide nanoparticles immobilized on activated carbon, which highlighted the effect of activated carbon as a practical active agent to increase the overall efficiency of the system.[17]

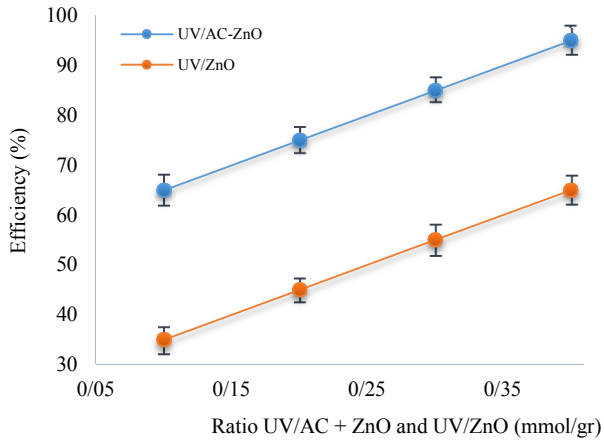


Fig. 9. Effect of changes in Zno alone and ZnO nanoparticles immobilized on activated carbon in removal of acid blue 113 dye (pH: 3, initial dye concentration: 100 mg/l, modified photocatalytic concentration: 100 mg/l, reaction time: 30 minutes)

Comparison of the efficiency of various processes in optimal conditions

At this stage, the optimum values of the parameters involved in the experiments were determined. At the optimum reaction time of 30 minutes, we investigated the effects of ultraviolet radiation, simultaneous use of ultraviolet and oxidizing radiation, and simultaneous effect of the zinc oxide nanoparticles immobilized on synthetic activated carbon in the presence of ultraviolet radiation. Figure 10 shows the efficiency of each process in terms of the overall system efficiency in optimum conditions. According to the obtained results, the removal efficiency of the process using the ultraviolet radiation, UV/ZnO process, and UV/AC+ZnO process was 20%, 65%, and 96%, respectively.[16, 32]

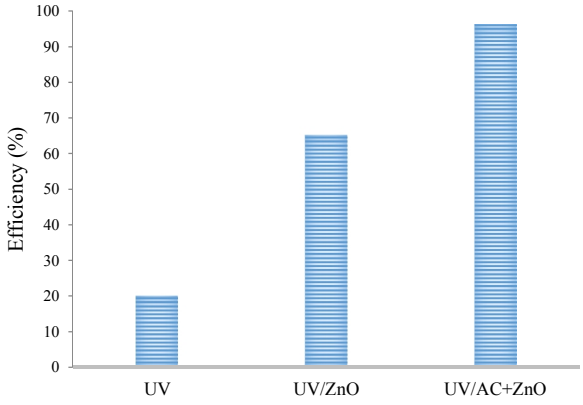


Fig. 10. Comparison of efficiency of various processes for acid blue 113 dye removal in optimum conditions (pH: 3, initial dye concentration: 100 mg/l, modified photocatalyst concentration: 100 mg/l, reaction time: 30 min)

Reaction kinetics

Chemical kinetics are used to examine the rate of chemical reactions. The reaction rate could be expressed by reducing the concentration of a reactant in a unit time or increasing the concentration of a product per unit time. The results of the present study in this regard are presented in Table 3 and Figures 11-13.

Based on the kinetic coefficients in Table 3, the removal rate in this process followed a first-order reaction since the correlation-coefficient in the first-order reaction (Figure 11) was higher than the correlation-coefficients in the zero- and second-order kinetics (figures 12 & 13). These findings are consistent with the results obtained by Muthirulan *et al.*, who reported that the reaction kinetics followed the first-order kinetics.[17]

Table 3. Kinetic coefficients of reaction in removal of acid blue 113 dye

NO	-Ln(C/C$_0$)	C/C$_0$	C (mg/L)	Time (min)	C$_0$ (mg/L)
1	100	5	65.91	0.659	0.416
2	100	10	36.67	0.366	1.003
3	100	15	27.20	0.272	1.301
4	100	20	13.71	0.137	1.986
5	100	30	3.18	0.031	3.449

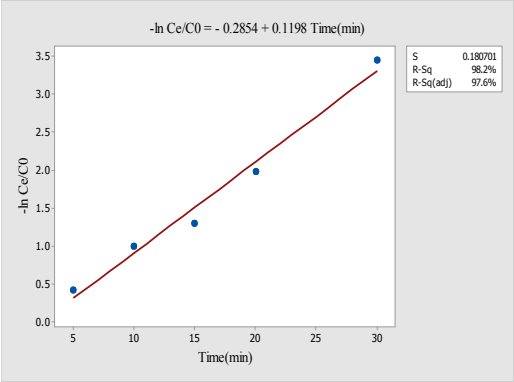


Fig. 11. First-order reaction kinetics for removal of acid blue 113 dye

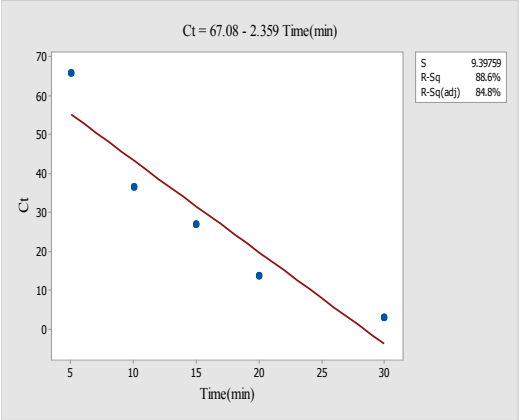


Fig. 12. Zero order reaction kinetic for removal of acid blue 113 dye

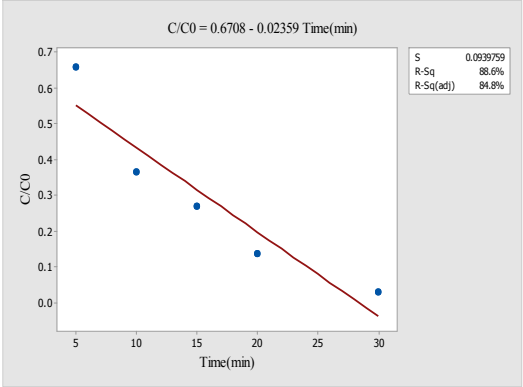


Fig. 13. Second-order reaction kinetic of removal of acid blue 113 dye

Conclusion

According to the results, there was a direct correlation between photocatalyst concentration and contact time with the process efficiency in the removal of the acid blue 113 dye. Moreover, the process efficiency was higher in acidic conditions compared to neutral and alkaline conditions. Our findings also indicated that the use of zinc oxide nanoparticles immobilized on synthetic activated carbon provided higher efficiency compared to the independent use of these materials.

Acknowledgements

This article was extracted from a research project approved by the School of Public Health at Hamadan University of Medical Sciences, Iran (code: 9610126532). Hereby, we extend our gratitude to the Deputy of Research and Technology for the support of this research project.

References

1. Seidmohammadi A, Asgari G, Leili M, Dargahi A, Mobarakian A. Effectiveness of quercus branti activated carbon in removal of methylene blue from aqueous solutions. Arch Hyg Sci 2015;4(4):217-25.
2. Shokoohi R, Ghovami Z, Dargahi A, Vanaee Tabar M. Evaluating the efficiency of ultrasonic and persulfate compilative process in Eosin Y dye removal from aquaeous solutions. J Color Sci Technol 2017;11(4):265-74.
3. Mahmoodi NM, Abdi J, Taghizadeh M, Taghizadeh A, Hayati B, Shekarchi AA, et al. Activated carbon/metal-organic framework nanocomposite: Preparation and photocatalytic dye degradation mathematical modeling from wastewater by least squares support vector machine. J Environ Manage 2019;233:660-72.
4. Forgacs E, Cserhati T, Oros G. Removal of synthetic dyes from wastewaters: a review. Environ Int 2004;30(7):953-71.
5. Pirsaheb M, Zinatizadeh, AK, Dargahi A. Performance evaluation of coagulation process in removal of low turbidity and color from water using different inorganic coagulants. J water & wastewater 2012;23(1):111-118.
6. Anliker R. Color chemistry and the environment. Ecotoxicol Environ Saf 1977;1(2):211-37.

7. Asgari G, Dargahi A, Mobarakian SA. Equilibrium and synthetic equations for index removal of methylene blue using activated carbon from oak fruit bark. J Mazandaran Univ Med Sci 2015;24(121):172-87.
8. Gupta V, Gupta B, Rastogi A, Agarwal S, Nayak A. A comparative investigation on adsorption performances of mesoporous activated carbon prepared from waste rubber tire and activated carbon for a hazardous azo dye—Acid Blue 113. J Hazard Mater 2011;186(1):891-901.
9. Mondal S. Methods of dye removal from dye house effluent—an overview. Environ Eng Sci 2008;25(3):383-96.
10. Raman CD, Kanmani S. Textile dye degradation using nano zero valent iron: a review. J Environ Manag 2016;177:341-55.
11. Shaban YA, El Sayed MA, El Maradny AA, Al Farawati RK, Al Zobidi MI. Photocatalytic degradation of phenol in natural seawater using visible light active carbon modified (CM)-n-TiO_2 nanoparticles under UV light and natural sunlight illuminations. Chemosphere 2013;91(3):307-13.
12. Soon AN, Hameed B. Heterogeneous catalytic treatment of synthetic dyes in aqueous media using Fenton and photo-assisted Fenton process. Desalination 2011;269(1-3):1-16.
13. Sulak MT, Yatmaz HC. Removal of textile dyes from aqueous solutions with eco-friendly biosorbent. Desalination Water Treat 2012;37(1-3):169-77.
14. Song S, Xu L, He Z, Ying H, Chen J, Xiao X, et al. Photocatalytic degradation of CI Direct Red 23 in aqueous solutions under UV irradiation using $SrTiO_3$/CeO_2 composite as the catalyst. J Hazard Mater 2008;152(3):1301-8.
15. Zollinger H. Color chemistry: syntheses, properties, and applications of organic dyes and pigments: John Wiley & Sons; 2003.
16. Shu H-Y, Chang M-C, Huang S-W. UV irradiation catalyzed persulfate advanced oxidation process for decolorization of Acid Blue 113 wastewater. Desalination Water Treat 2015;54(4-5):1013-21.
17. Muthirulan P, Meenakshisundararam M, Kannan N. Beneficial role of ZnO photocatalyst supported with porous activated carbon for the mineralization of alizarin cyanin green dye in aqueous solution. J Adv Res 2013;4(6):479-84.
18. Pandiyarajan T, Mangalaraja RV, Karthikeyan B, Sathishkumar P, Mansilla HD, Contreras D, et al. UV-A light-induced photodegradation of Acid Blue 113 in the presence of Sm-doped ZnO nanostructures. Appl Phys A 2015;119(2):487-95.
19. Ahmad A, Hameed B. Fixed-bed adsorption of reactive azo dye onto granular activated carbon prepared from waste. J Hazard Mater 2010;175(1-3):298-303.
20. Fen L, Bo Y, Wei J, Zhenlong G. Preparation of Activated Carbon Loading Nano-ZnO and Desulfurization Properties under Room Temperature. In: Zhang W. (eds) Software Engineering and Knowledge Engineering: Theory and Practice. Advances in Intelligent and Soft Computing, 2012; 162: Berlin, Heidelberg.
21. Ghaedi M, Sadeghian B, Pebdani AA, Sahraei R, Daneshfar A, Duran C. Kinetics, thermodynamics and equilibrium evaluation of direct yellow 12 removal by adsorption onto silver nanoparticles loaded activated carbon. Chem Eng J 2012;187:133-41.
22. Kataria N, Garg V. Removal of Congo red and Brilliant green dyes from aqueous solution using flower shaped ZnO nanoparticles. J Environ Chem Eng 2017;5(6):5420-8.
23. Arbabi M, Mayahi B, Mohammadi Moghadam F, Sedehi M, Hemati S. Removal of acid bleu 113 by UV/H_2O_2/Fe_3O_4 process: Optimization of treatment conditions using experimental design. J Shahrekord Univ Med Sci 2017;19.
24. Mehralipour J, Dayari A, Rezaei Vahidian H, Samarghandi M, Azizi F. Optimization of electro-proxone via response surface statistical model in decolorization of acid orange 7 dye from synthetic wastewater. J Color Sci Technol 2016;10(4):247-58.
25. Kadirova ZC, Katsumata K-i, Isobe T, Matsushita N, Nakajima A, Okada K. Adsorption and photodegradation of methylene blue by iron oxide impregnated on granular activated carbons in an oxalate solution. Appl Surf Sci 2013;284:72-9.
26. Kousha M, Daneshvar E, Dopeikar H, Taghavi D, Bhatnagar A. Box–Behnken design optimization of Acid Black 1 dye biosorption by different brown macroalgae. Chem Eng J 2012;179:158-68.
27. Hadi M, Samarghandi MR, McKay G. Equilibrium two-parameter isotherms of acid dyes sorption by activated carbons: study of residual errors. Chem Eng J 2010;160(2):408-16.
28. Samarghandi MR, Siboni M, Maleki A, Jafari SJ, Nazemi F. Kinetic Determination and efficiency of titanium dioxide photocatalytic process in removal of reactive Black 5 (RB5) dye and

cyanide from aquatic solution. J Mazandaran Univ Med Sci 2011;21 (81): 44-52.
29. Mehrizad A, Gharbani P. Removal of methylene blue from aqueous solution using nano-TiO2/UV process: optimization by response surface methodology. Prog Color Colorants Coat 2016;9:135-43.
30. Rahmani AR, Zamani F, Shabanloo A, Almasi H. Effect of silica on the ultrasonic/persulfate process for degradation of Acid Black 1 in aqueous solutions. Avicenna J Environ Health Eng 2016;3(2).
31. Seid-Mohammadi A, Shabanloo A, Fazlzadeh M, Poureshgh Y. Degradation of acid blue 113 by US/H_2O_2/Fe^{2+} and US/$S_2O_8^{2-}$/Fe^{2+} processes from aqueous solutions. Desalination Water Treat 2017;78:273-80.
32. Hunger K. Industrial dyes: chemistry, properties, applications: John Wiley & Sons; 2007.