Biological Environment and Pollution
Vol. 1(2) 2021 | 76 – 88
ISSN 2798-2378
ISSN 2798-2777

Degradation of AB113 using combined photocatalysis and biological processes: Economic choice and enhanced removal of aromatic compounds

Roya Nayebi Gavgani [1], Bita Ayati [2,*], Hossein Ganjidoust [3]
Environmental Eng. Division, Civil & Environmental Eng. Faculty, Tarbiat Modares University, Tehran, Iran
[1] nayebi.roya@gmail.com ; [2] ayati_bi@modares.ac.ir; [3] h-ganji@modares.ac.ir
* Corresponding author

ARTICLE INFO

Article history
Submission
September 18, 2021
Revision
November 28, 2021
Accepted
December 21, 2021

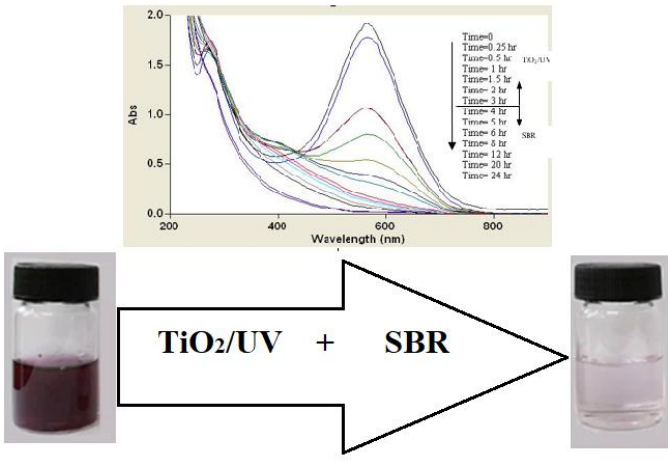
Keyword
Photocatalytic
Biological
Economic
Kinetic

ABSTRACT

Dyes are important chemical compounds which are applied to textile, paper, food, printing, leather, cosmetics industries, and so on. Textile dyes are aromatic and hardly degradable compounds causing harmful effects such as toxicity, carcinogenesis, and mutagenesis. Conventional treatment processes such as biological processes, or adsorption to activated carbon are not very effective for textile wastewater treatment, and therefore advanced treatment process was extended. In this study, decolorization and mineralization of Acid Blue 113 were investigated using combined TiO_2/UV-SBR systems. Dye concentration in the effluent of photocatalytic reactor was selected via efficiency of dye removal, BOD_5/COD ratio, and energy consumption of alkali media. Three hours after applying the samples to the photocatalytic reactor, more than 80% of the dye was removed and the rest was decolorized approximately after passing bioreactor. Investigation of optimum condition leads to enhance the degradation of aromatic compounds. The results show that second order and Grau are the best models to describe the photocatalytic and biological kinetic data with higher correlation coefficients, respectively.

Conflict of interest: The authors declare that they have no conflicts of interest.

GRAPHIC ABSTRACT



Introduction

Approximately, 100,000 commercially available dyes and pigments are used in industries such as textile dyeing, cosmetics, and paper production, and around 700,000 tons of synthetic dyes are produced annually. Researchers report that about 10-15% of consuming dyes are lost in the effluent during dyeing processes[1,2]. The wastewater emerging from textile industry has the lowest ratio of BOD_5: COD (<0.1)[3,4]. Dyes change the color of water and prevent light diffusion, disturb photosynthesis and lead to destruction of aquatic ecosystem[1,5]. Modern commercial dyes are very stable and contain a high composition ratio of aromatic rings[6]. Azo dyes are the most applied class of organics. The release of these compounds into the environment is undesirable; not only because of their color, but also because their breakdown products are toxic and/or mutagenic to life. Thus, wastewater containing synthetic azo dyes is considered a serious warning for the environment and removal of these pollutants has been attended in several researches[6–8]. However, conventional treatment techniques including physical, chemical, and biological, such as adsorption, filtration, coagulation, and chemical oxidation are unable to provide environmental standards of degradation[1,9]. Advanced oxidation processes (AOPs) such as ozonation, electrochemical oxidation and photocatalytic methods are proper to mineralize dye wastewater[10].

In the hybrid methods toxic and non-degradable chemicals endure chemical process, breakdown to compounds with less toxic nature and more biodegradable. For example, coupling both biological and chemical processes could be an efficient approach to decrease the cost of wastewater treatment, energy consumption, and complete degradation time[11,12]. Recently, many coupled systems such as up-flow anaerobic sludge blanket (UASB) reactor[13] and ozonation, Fenton's reagent with aerobic biological treatment, and ozonation[14,15] with biological treatment[11] have been studied to treat different kinds of industrial wastewaters such as effluents from textile industry, dye house liquors, olive mill, and polyester resin production.

In recent years, application of advanced oxidation methods (AOPs) such as photocatalytic reactions have been expanded for removing non-biodegradable and toxic pollutants[16,17]. When photo-catalyst such as titanium dioxide (TiO_2) absorbs ultraviolet radiation from sunlight or illuminated light source, it will produce pairs of electrons and holes. The electron of the valence band becomes excited when illuminated by light. Also, the excess energy of this excited electron promoted the electron to the conduction band. The generated holes oxidize the organic compounds and produce positive charged molecules (R^+), which would react with OH^- and finally the hydroxide radical is generated[18]. Also, photocatalytic process gradually breaks down the contaminant molecules under room temperature and pressure, no residue of the original material remains and therefore no sludge requiring disposal to landfill is produced[19].

In the past two decades, significant studies have been done using microorganisms to remove environmental contaminants from textile wastewater. Different microorganisms such as aerobic and anaerobic bacteria and fungi could degrade azo dyes[20,21]. Sequencing batch reactor (SBR) is a wastewater treatment process based on the activated sludge system. The operating cycle is divided into five phases, including filling, aeration-reaction, settling, decanting and idle. With respect to the application, SBR has been successfully employed in biodegradation of both municipal and industrial wastewater[22,23].

Wang and his colleagues worked on Reactive Black (RB5) solution degradation using coupled electrochemical/photocatalyst degradation processes. Rapid decolorization rate of RB5 solution (55 and 110 µmol/dm^3) was yielded by applying the electrochemical oxidation method at pH 4 and current density of 277 A/m^2 with enough electrolytes (2 g NaCl/dm^3). In addition, more than 95% of dye was removed for 4 minutes. In photocatalytic method, 63% and 23% of RB5 was degraded by immobilized TiO_2 within 5 hours for initial dye concentration of 55 and 110 µmol/ dm^3, respectively. By combining the two methods, the researchers concluded that the concentration of the dye and TOC was effectively reduced[24]. In another study, Chengyuan

and his coworkers[25] studied effect of cephalexin after heterogeneous Fenton-like pretreatment on the performance of anaerobic granular sludge and activated sludge. They concluded that Fe_3O_4 NPs Fenton-biotreatment was an effective approach in treating cephalexin. Also, Humic acid in LB-EPS and TB-EPS decreased after pretreatment and Rhodobacteraceae have the most significant increase at pretreatment condition[25].

Acid dyes which are organic sulfonic acids with high solubility are the most widely used chemicals in the industry for dying different fibers such as cotton, rayon, and polyester[6]. Currently, these types of dyes are increasingly used in I.R. Iran. According to the statistics reports, 141000 tons of acid dyes were imported in 2011[26].

The main objective of the present study was to investigate the performance of immobilized photocatalytic reactor on concrete surface as a pre-treatment unit to enhance dye removal by breakdown of aromatic rings. For this reason, the combined photocatalytic–biological process was studied for dye degradation, which not only makes it possible but also was more economical considering the high costs of using merely chemical processes. The effects of variables, including initial pH, dye concentration, energy consumption, irradiation intensity and BOD_5/COD ratio were studied by changing each in turn while keeping the other parameters constant.

Method

Material properties

Acid Blue 113 (AB113) (molecular weight=681.6 g/mol, solution pH=6-9, chemical structure: diazo) was purchased from Alvan Sabet Company (Tehran, Iran) and used without any purification (Figure 1). The photocatalyst was P25 titania from Degussa Company (density=5 g/cm^3, particle size=21 nm, ratio of anatase to rutile=80:20). All the materials for biological system feeding were in analytical grade (Merck, Germany). Also, K_2CrO_7, $AgSO_4$, $HgSO_4$, and NaOH (Merck, Germany), were used for the COD measurement. UV-C lamps were purchased from Philips Company.

The applied spectrophotometer was Carry 50, Varian Co. The COD reactor and BOD meter which have been used for determining the amounts of organic pollution were DRB200, Hatch Co. and OxiTop, WTW Co., respectively. Also, ultrasound was used for dispersing nano particles (Fungilab, UE-6SFD) and H_2SO_4 and NaOH were employed for pH adjustment in the range of 3-11. In addition, pH meter (Metrohm, 691) and centrifuge (CE145, Shimi Fan Co.) have been applied.

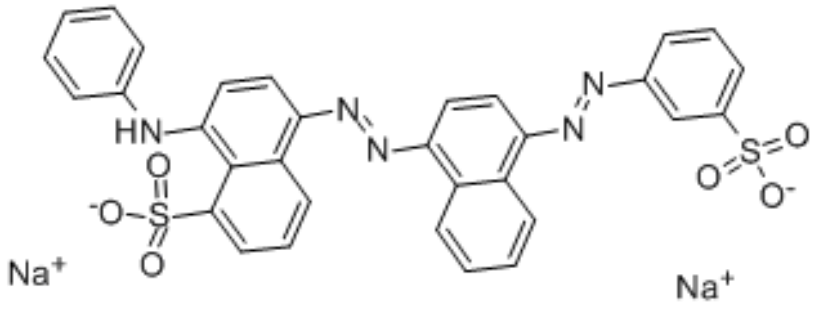


Fig 1. Structure of acid blue 113

Configuration of combined system

The combined lab-scale photocatalytic–SBR system used for the mineralization of bio-recalcitrant AB113 has been designed and constructed in the environmental engineering laboratory (Figure 2). The photocatalytic reactor was a plastic container (length=0.23 m, width=0.16 m, height=0.05 m), inside which a concrete bed with 0.008 m thickness was located.

An immobilized (TiO_2=40 g/m^2) system was applied by concrete glue[27]. To reach the maximum percentage of UV irradiation reflection and safety aspects, inner and outer surfaces of the reactor were covered with aluminum foil. To investigate the effect of UV-C irradiation intensity, the UV-C lamps with power of 30, 90, 150 and 210 Watts were used (for instance, symbol P_{UVC}=30 W means UV-C lamp with 30 Watts power). Total volume of dye solution was 25 cm^3 in each reactor. The bioreactor consists of a sequencing batch reactor made of Plexiglas with inner diameter of 0.1 m, height of 0.7 m, and 3 L effective volumes.

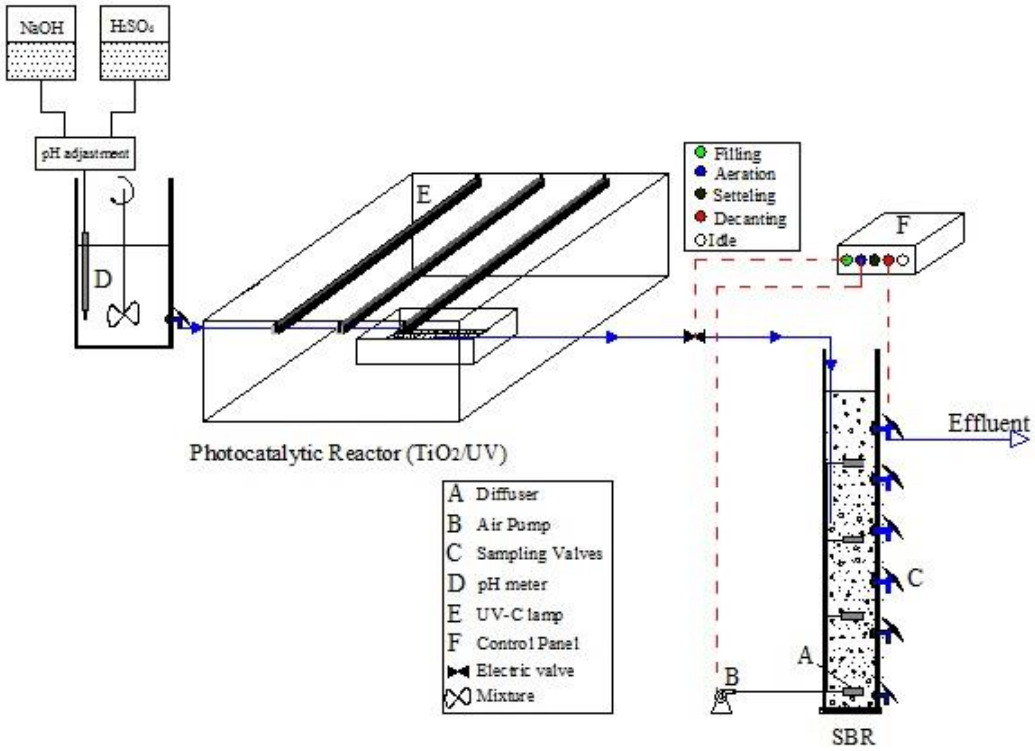


Fig 2. Schematic of the experimental photocatalytic–biological reactors used in this study

Composition of synthetic wastewater

The synthetic wastewater for photocatalytic reactor was prepared with different concentration of AB113 including 50, 75, 100, 150 and 200 mg/L in tap water. C and C_0 were used as a dye concentration and initial dye concentration, respectively.

A mineral nutrients solution was added separately to the bioreactor that included nitrogen and phosphorus source ($MgSO_4.7H_2O$, $FeCl_3.6H_2O$ and $Na_2MoO_4.2H_2O$) to keep the C/N/P ratio at 100:5:1. The sludge was prepared from sanitary wastewater (Shahrak Gharb wastewater treatment plant, Tehran, Iran). The adaptation period lasted 120 days (the initial COD=200 mg O_2/L). During this time, a combination of dye and glucose was used as feed. Also, a 10% increase of dye followed by the same decrease in glucose amount was done in each step of system feeding. In the main experiments, only dye was used as feed for the reactors. After the adaptation step, dye concentration was gradually increased to 200, 250, 300, 400 and 500 mg/L. The loading repetition was continued during each increase in dye concentration until reaching a stable state. The changes in COD removal were trivial because of the adaptation of microorganisms with the dye.

Also, during the operation period, mixed liquor suspended solids (MLSS) and sludge volume index (SVI) reached a nearly constant amount of 1837±339 mg/L and 68±12 cm^3/g of MLSS, respectively (Figure 3).

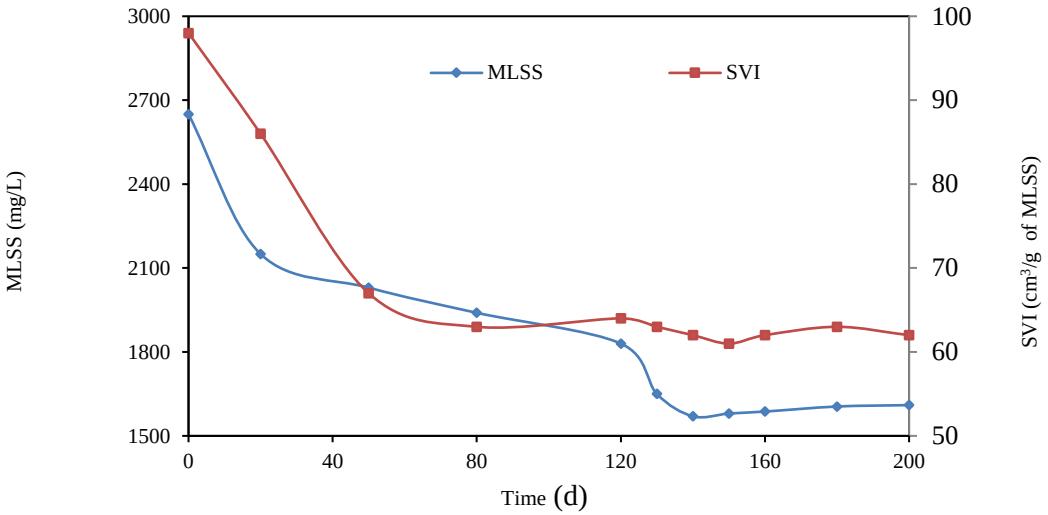


Fig 3. Changes in MLSS and SVI in SBR system

At the beginning of the adaptation period, because of the difficulty of dye degradation, the amount of MLSS and SVI decreased with time, and then tended to gradually increase. The variations in both MLSS and SVI were the same. The characteristics of the SBR system for different concentrations of dye are shown in Table1.

Table 1. Time ranges of SBR system for different (COD_{total}=200 mg O_2/dm_3)

Time (min)	**COD_{dye}/COD_{total}**		
Stage	**Less than 0.75**	**0.25 - 0.75**	**More than 0.25**
Filling	2	2	2
Aeration-reaction	180	420	1380
Settling	20	20	20
Decanting	8	8	8
Idle	30	30	30

Analytical methods and procedures

Dye concentration was determined by measuring the absorbance of the test samples at the maximum absorbance wavelength of AB113 (λ_{max}:566 nm) using UV–Vis spectrophotometer. Before the analysis, samples withdrawn from the treatment systems were centrifuged at 3000 rpm for 10 min. According to the standard methods[27], COD and BOD_5 analysis was carried out with COD and BOD reactor. BOD was measured by standard method 5210 B (5-day BOD test) according to Sawyer and McCarty[27]. The BOD measurement with OxiTop® is based on pressure measurement in a closed system: The micro-organisms in the sample consume the oxygen and generate CO_2 in the process. This is absorbed with NaOH. This will cause a vacuum pressure, which can be determined as a measured value in mg/L BOD. All experiments were conducted up to three times at room temperature (22 ± 1°C).

Results and Discussion

Effect of initial pH on TiO_2/UV process

The initial pH is one of the effective parameters influencing TiO_2/UV process that is shown in Figure 4. According to the results, 33.4, 51.9, 81.8 and 84.1% of AB113 was removed in 5 hours at pH 3, 6.5 (neutral), 9 and 11, correspondingly. The optimum pH was observed in the alkali condition due to more and reliable generation of hydroxide radicals[19,28].

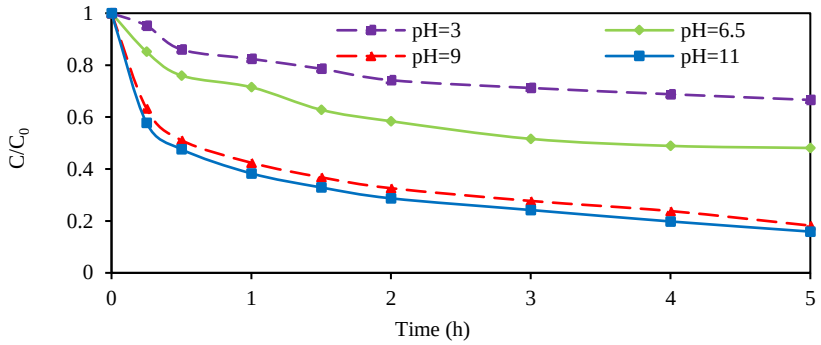


Fig 4. Effect of initial pH on the decolorization of AB113 solution by TiO_2/UV (TiO_2=40 g/m^2, C_0=50 mg/L, P_{UVC}=30 W)

In pH = 3, adsorption of dye molecules onto TiO_2 surface has an important role in dye removal because of the difference in surface charge between TiO_2 particles and dye molecules (PZC of TiO_2 is at pH 6.8)[29]. At the first hours, suitable removal efficiency was observed at pH=3, which can be explained by more effect of OH^0 as compared to hole oxidizing. Also, changes of pH were monitored during the process to justify possibly generated intermediates[30]. In this research, the pH of different media (alkali, neutral, and acidic) was trended to neutral during the process time (Figure 5). The optimum pH was found to be around 9 by considering dye removal efficiency, the energy costs, NaOH consumption and effluent pH.

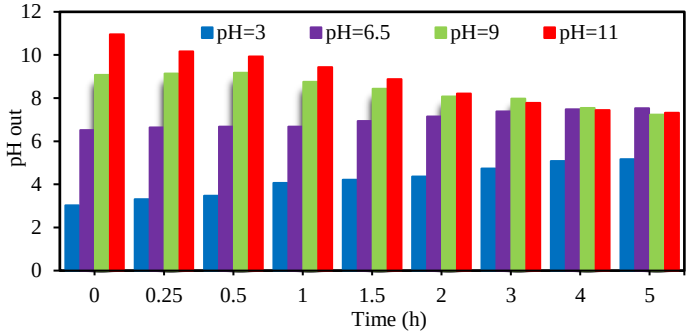


Fig 5. Changes of pH during TiO2/UV reaction (TiO_2=40 g/m^2, C_0=50 mg/L, P_{UVC}=30 W)

Effect of UV-C intensity on TiO_2/UV process

According to the results, dye removal increased in high irradiation intensity (Figure 6). Numbers of emitted photons were enhanced by increasing intensity of UV irradiation. But, when UV-C irradiation intensity was more than special value, the removal efficiency remained constant. The reason was that this intensity was equal to maximum energy for exciting immobilized TiO_2 on a concrete bed. Thus, 90 Watts was selected as the optimum UV-C irradiation intensity.

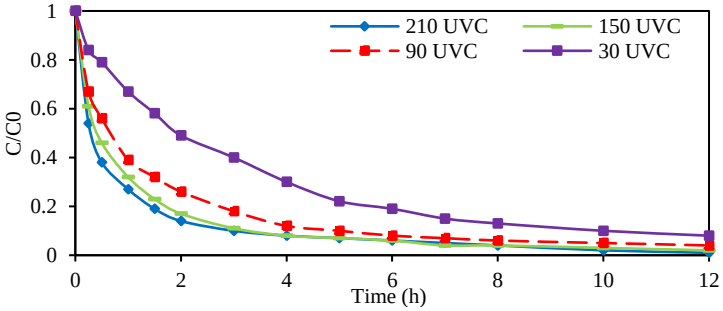


Fig 6. Effect of UV-C irradiation intensity on the decolorization of AB113 solution by TiO_2/UV (TiO_2=40 g/m^2, pH=9, C_0=50 mg/L)

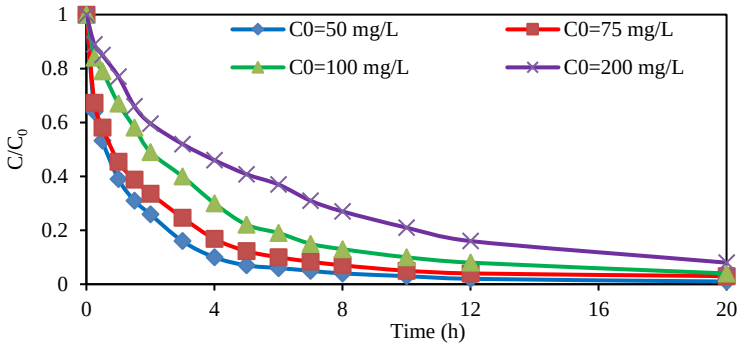


Fig 7. Effect of initial dye concentration on the removal of AB113 by TiO_2/UV (TiO_2=40 g/m^2, pH=9, P_{UVC}=90 W)

Effect of initial dye concentration on TiO_2/UV process

According to Figure 7, 83.4, 77.9, 59.2 and 42.8% of AB113 was removed in 5 hours when dye concentration amounted to 50, 75, 100 and 200 mg/L, respectively. At high concentrations, more dye was adsorbed onto the TiO_2 surface. Consequently, the UV irradiation into the dye particle was hindered. In addition, by increasing the concentration of the dye, active sites onto the nano particles surface would be more occupied. Also, a significant amount of UV was absorbed by the dye molecules rather than TiO_2 particles[19].

According to what was mentioned above, the concentration of $OH^{\bullet}$ was decreased by increasing the concentration of the dye. Therefore, the efficiency of the catalytic reaction was reduced[19]. The presumed reason was that some photocatalytic reactions occurred, which caused degradation and removal of intermediates[31,32]. Whereas dye removal in a short time could not be a logical selection for optimum concentration and effluent of photocatalytic reactor discharge to a biological system, then COD/COD_0, BOD_5/COD and energy consumption were investigated. As is evident from (Figure 8), the COD/COD_0 decreased at the first 20 min which can be contributed to the breaking down of N=N bond, and then it was enhanced due to degradation and simplification of the available compounds. In the following, oxidants are expected to substantially improve the degradation rates of products and as a result the rate of COD/COD_0 decreased during the treatment process. In addition, BOD_5/COD was measured at inflection and pick points (Figure 9). According to Equation (1)[33], the amount of energy consumption could be calculated to choose the suitable concentration based on influent BOD/COD in SBR.

$$EC/C=(W.t)/(C_{in}-C_{out}) \quad (1)$$

Whereas W is energy consumption kW, t shows the time of radiation in hour, EC/C is the used energy for removal of pollutant in kWh.L/mg. C_{in} and C_{out} are the initial and final concentration in mg/L after photocatalytic process, respectively.

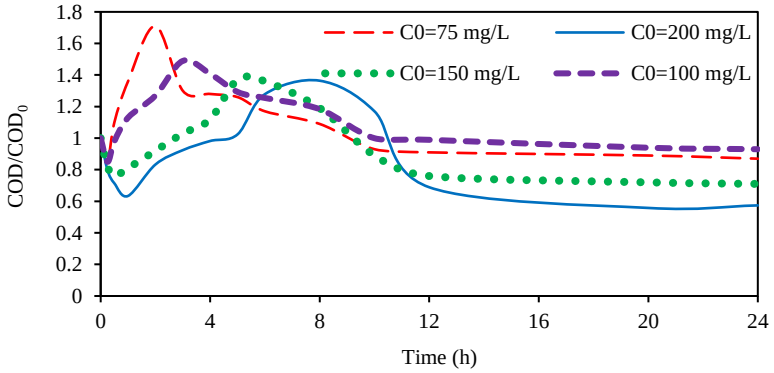


Fig 8. COD removal for different initial concentration of AB113 by TiO_2/UV (TiO_2=40 g/m^2, pH=9, P_{UVC}=90W)

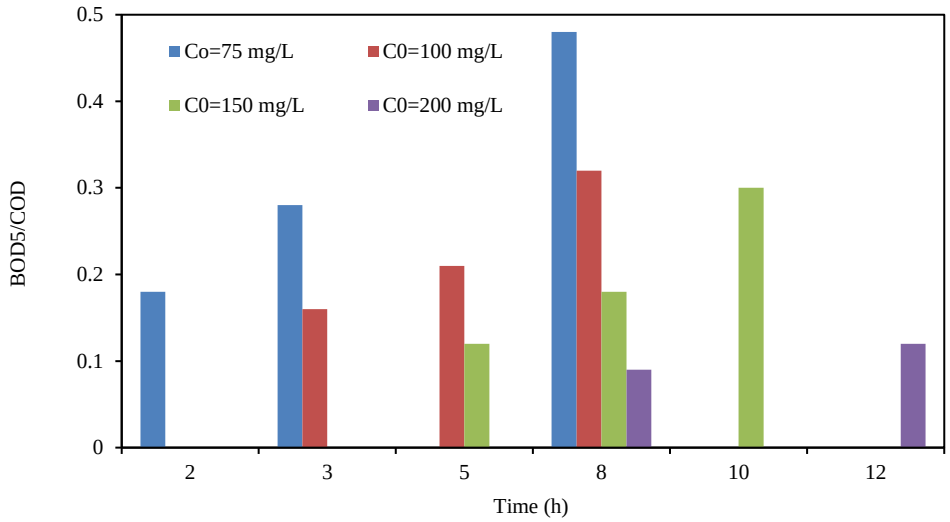


Fig 9. BOD_5/COD ratio for different initial concentration of AB113 in TiO_2/UV process (TiO_2=40 g/m^2, pH=9, P_{UVC}=90 W)

According to Figure 10, the amounts of 4.42, 9.35 and 7.8 Wh. L/mg were obtained for 75, 100 and 150 mg/L initial dye concentrations for the same BOD_5/COD (around 0.4), respectively. Thus, after 3 hours 75 mg/L dye was added to the bioreactor.

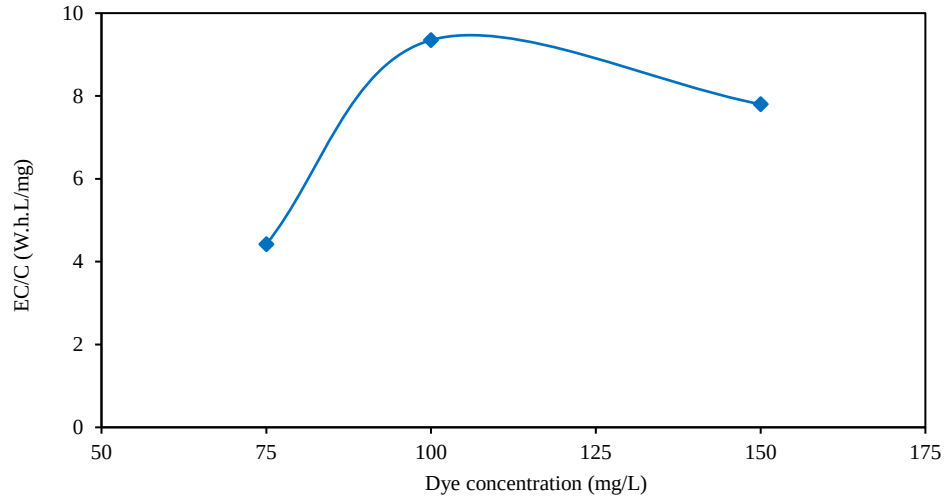


Fig 10. Energy consumption for different initial concentration of AB113 in TiO_2/UV process (TiO_2=40 g/m^2, pH=9, P_{UVC}=90 W)

Capability investigation of combined system

Figure 11, Tables 2 and 3 show the absorption spectra for synthetic wastewater in the combined system and both photocatalytic and biological systems influent and effluent characteristics, respectively. There was 80% decolorization in the photocatalytic reactor after 3 hours. The residual color was completely removed, and the COD value was significantly diminished in the biological reactor. Also, the COD, BOD_5 and BOD_5/COD values increased in the effluent. This means that the dye complex molecules have been degraded and can be treated by the biological system. According to the results, the effluent from SBR has the environmental standard discharge concentration after about 20 hours.

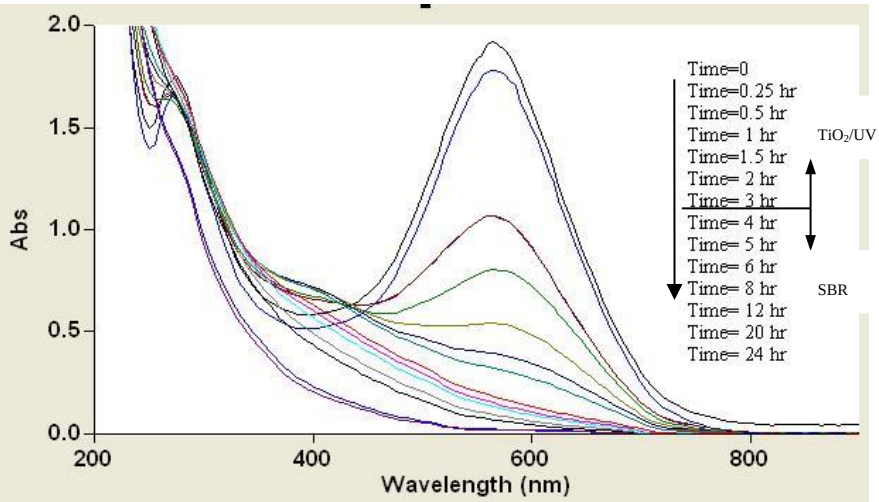


Fig 11. Absorption spectra of synthetics wastewater in different time (pH=9, C_0=50 mg/L)

Table 2. Influent and effluent parameters of photocatalytic reactor after 3 hours

Parameter	Influent	Effluent
Dye concentration (mg/L)	75	13.4
COD (mg O_2/L)	136.2	176.8
BOD_5 (mg O_2/L)	10	50
BOD_5/COD	0.08	0.4

Table 3. Influent and effluent parameters of bioreactor after 20 hours

Parameter	Influent	Effluent
Dye concentration (mg/ L)	13.4	0.3
COD (mg/L)	176.8	52.3
BOD_5 (mg/L)	50	25
MLSS (mg/L)	1837±339	
MLVSS [1](mg/L)	1507±273	
MLVSS/MLSS	82.1±1.3	
SVI (cm^3/g of MLSS)	68±12	

Investigation of AB113 degradation

Decolorization is not the actual degradation of dye, since breaking specific bonds in molecule causes decolorization. Thus, after optimizing the parameters, in order to have more accurate consideration, UV-Vis absorption was determined at wavelengths 566, 255, 276[34], and 271 nm which were related to the known compounds AB113, benzene, naphthalene, and phenol, respectively[28,35]. As shown in Figure 12, after 3 hours (effluent of photocatalytic reactor), removal of AB113, benzene, naphthalene, and phenol were 82.1, 11.3, 5.4, and 6.6% percent, respectively. Also, after 20 hours (effluent of bioreactor), those ratios reached 99.3, 27.2, 19.7 and 22.4 %, respectively. The presumed reason for increasing the removal of aromatic compounds is their degradation by adapted microorganisms or adsorption onto sludge due to hydrophobic structure.

[1] Mixed Liquor Volatile Suspended Solids

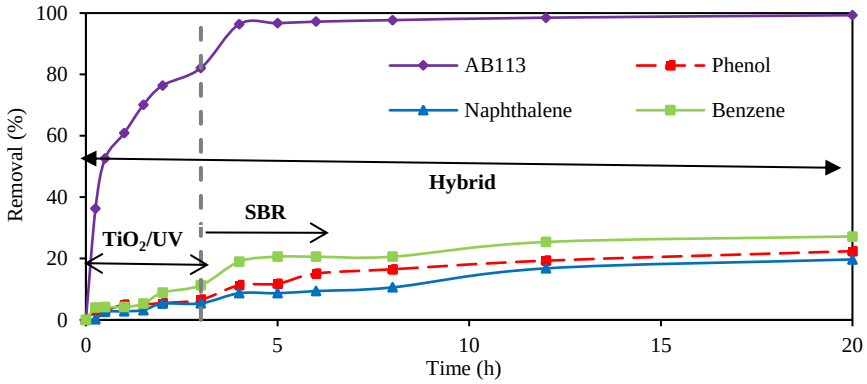


Fig 12. Comparison of removal between dye, phenol, naphthalene, and benzene (pH=9, C_0=50 mg/L)

Kinetic study

Reactions in a photocatalytic reactor were categorized as zero-order, Langmuir-Hinshelwood (first-order) and the second-order reactions[36]. Considering the optimum pH at 9, PZC of TiO_2 (at pH=6.8) and anionic structure of the dye molecule, it was observed that in this case absorbance was not the efficient factor. According to the results (Figure 13), the second-order kinetic of reaction had a decent agreement with the experimental data (R^2=0.989).

Three kinetic models such as first-order, Grau and Stover–Kincannon have been also applied for the biological reactors[37,38]. According to the results, the decolorization kinetic was found to follow Grau model with a higher correlation coefficient of 99.8 %. In Grau model, θ_H, E are hydraulic retention times (h) and removal rate, respectively.

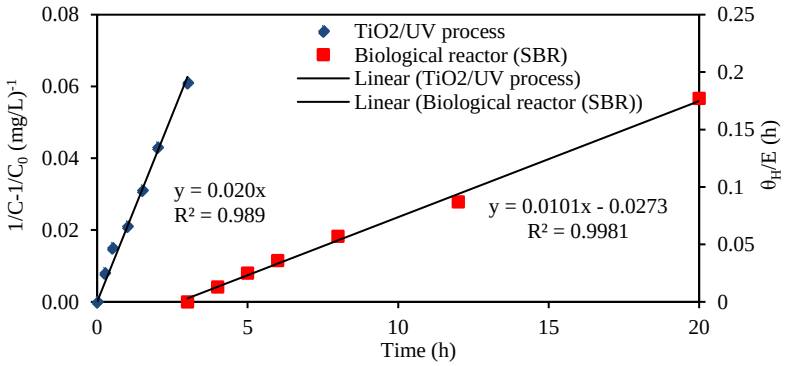


Fig 13. Graphic representation of kinetic models (TiO_2=40 g/m^2, C_0=75 mg/L, pH=9, P_{UVC}=90 W, MLSS=1600 mg/L, DO[2]=2.8 mg/L)

Conclusion

Since the biological systems have limitations for dye removal, using combined systems is essential. The feasibility of using a combined method involving photocatalyst, followed by the biological treatment for dye removal was confirmed. According to the results, dye concentration in the effluent of photocatalytic reactor (75 mg/L) was selected via BOD_5/COD (0.4) ratio and energy consumption in alkali media. Furthermore, more than 80 % of the dye was removed after 3 hours and the residual color was removed after passing through bioreactor. Also, second order and Grau models were selected as the kinetic model for photocatalytic and biological reactors, correspondingly.

[2] Dissolved Oxygen

Acknowledgment

The authors are thankful for the INIC (Iran Nanotechnology Initiative Council) financial support ofthis research andalso, to Dr. Yaghoub Fathollahi, the Vice-Chancellor for Research Affairs of Tarbiat Modares University. The valuable supports of Environmental Engineering Laboratory of Civil and Environmental Faculty of Tarbiat Modares University are also appreciated.

References

1. Gregorio, S. D. *et al.* Sustainable Discoloration of Textile Chromo-Baths by Spent Mushroom Substrate from the Industrial Cultivation of Pleurotus ostreatus. *Journal of Environmental Protection* **1**, 85–94 (2010). https://doi.org/10.4236/jep.2010.12011
2. Samarghandi, M. R. *et al.* Application of acidic treated pumice as an adsorbent for the removal of azo dye from aqueous solutions: kinetic, equilibrium and thermodynamic studies. *Iranian J Environ Health Sci Eng* **9**, 9–18 (2012). https://doi.org/10.1186/1735-2746-9-9
3. Metcalf & Eddy. *Wastewater engineering : treatment and reuse*. (McGraw-Hill, 2003).
4. Kalra, S. S., Mohan, S., Sinha, A. & Singh, G. Advanced oxidation processes for treatment of textile and dye wastewater: A review. *International Conference on Environmental Science and Development IPCBEE* **4**, 271–275 (2011).
5. Ahmed, M. J. & Dhedan, S. K. Equilibrium isotherms and kinetics modeling of methylene blue adsorption on agricultural wastes-based activated carbons. *Fluid Phase Equilibria* **317**, 9–14 (2012). https://doi.org/10.1016/j.fluid.2011.12.026
6. Itodo, A. U., Usman, A., Akinrinmade, G., Itodo, H. U. & Ugboaja, V. C. Performance assessment of received and formulated carbon animalis: a comparative adsorption isotherm test. *Journal of Environmental Protection* **3**, 288–295 (2012). https://doi.org/10.4236/jep.2012.33036
7. Holkar, C. R., Jadhav, A. J., Pinjari, D. V., Mahamuni, N. M. & Pandit, A. B. A critical review on textile wastewater treatments: Possible approaches. *Journal of Environmental Management* **182**, 351–366 (2016). https://doi.org/10.1016/j.jenvman.2016.07.090
8. Carmen, Z. & Daniela, S. *Textile organic dyes – characteristics, polluting effects and separation/elimination procedures from industrial effluents – a critical overview. Organic Pollutants Ten Years After the Stockholm Convention - Environmental and Analytical Update* (IntechOpen, 2012). doi:10.5772/32373. https://doi.org/10.5772/32373
9. Peydayesh, M. & Rahbar-Kelishami, A. Adsorption of methylene blue onto Platanus orientalis leaf powder: Kinetic, equilibrium and thermodynamic studies. *Journal of Industrial and Engineering Chemistry* **21**, 1014–1019 (2015). https://doi.org/10.1016/j.jiec.2014.05.010
10. Amiri, H., Ayati, B. & Ganjidoust, H. Textile dye removal using a photocatalytic cascade disc reactor coated by ZnO nanoparticles: effects of hydraulic parameters. *Journal of Environmental Engineering* **142**, 04016019 (2016). https://doi.org/10.1061/(ASCE)EE.1943-7870.0001092
11. Kanagaraj, J. & Mandal, A. B. Combined biodegradation and ozonation for removal of tannins and dyes for the reduction of pollution loads. *Environ Sci Pollut Res Int* **19**, 42–52 (2012). https://doi.org/10.1007/s11356-011-0534-0
12. Gosavi, V. & Sharma, S. A general review on various treatment methods for textile wastewater. *Journal of Environmental Science, Computer Science and Engineering & Technology* **3**, 29–39 (2013).

13. Shankar, R. *et al.* Chapter 9 - Biological wastewater treatment plants (WWTPs) for industrial wastewater. in *Microbial Ecology of Wastewater Treatment Plants* (eds. Shah, M. & Rodriguez-Couto, S.) 193–216 (Elsevier, 2021). https://doi.org/10.1016/B978-0-12-822503-5.00023-0
14. Gunawan, D., Kuswadi, V., Sapei, L. & Riadi, L. Yarn dyed wastewater treatment using hybrid electrocoagulation-Fenton method in a continuous system: Technical and economical viewpoint. *Environmental Engineering Research* **23**, 114–119 (2017). https://doi.org/10.4491/eer.2017.108
15. Li, B. & Zhang, T. Removal mechanisms and kinetics of trace tetracycline by two types of activated sludge treating freshwater sewage and saline sewage. *Environ Sci Pollut Res* **20**, 3024–3033 (2013). https://doi.org/10.1007/s11356-012-1213-5
16. Shokrollahzadeh, S., Abassi, M. & Ranjbar, M. A new nano-ZnO/perlite as an efficient catalyst for catalytic ozonation of azo dye. *Environmental Engineering Research* **24**, 513–520 (2019). https://doi.org/10.4491/eer.2018.322
17. Trojanowicz, M., Bojanowska-Czajka, A., Bartosiewicz, I. & Kulisa, K. Advanced Oxidation/Reduction Processes treatment for aqueous perfluorooctanoate (PFOA) and perfluorooctanesulfonate (PFOS) – A review of recent advances. *Chemical Engineering Journal* **336**, 170–199 (2018). https://doi.org/10.1016/j.cej.2017.10.153
18. Ibhadon, A. O. & Fitzpatrick, P. Heterogeneous photocatalysis: recent advances and applications. *Catalysts* **3**, 189–218 (2013). https://doi.org/10.3390/catal3010189
19. Butler, E., Hunt, E., Fu, Y.-P., Mulamba, O. & Spaar, N. Investigating the governing decolorization mechanisms of nanodiamond in the treatment of an azo dye. *Desalination And Water Treatment* **86**, 183–190 (2017). https://doi.org/10.5004/dwt.2017.21252
20. Mani, A. & Hameed, S. A. S. Improved bacterial-fungal consortium as an alternative approach for enhanced decolourisation and degradation of azo dyes: a review. *Nature Environment and Pollution Technology* **18**, 49–64 (2019).
21. Mehrali, S., Moghaddam, M. R. A. & Hashemi, S. H. Feasibility study of several cyclic anaerobic/aerobic conditions in SBR system for treating of simulated dye (reactive blue19) wastewater. *Environ. Eng. Manag. J.* **11**, 617–621 (2012). https://doi.org/10.30638/eemj.2012.077
22. Elmolla, E. S. & Chaudhuri, M. The feasibility of using combined TiO2 photocatalysis-SBR process for antibiotic wastewater treatment. *Desalination* **272**, 218–224 (2011). https://doi.org/10.1016/j.desal.2011.01.020
23. Hameed, B. B. & Ismail, Z. Z. Biodegradation of reactive yellow dye using mixed cells immobilized in different biocarriers by sequential anaerobic/aerobic biotreatment: experimental and modelling study. *Environmental Technology* **42**, 2991–3010 (2021). https://doi.org/10.1080/09593330.2020.1720306
24. Chau, J. H. F. *et al.* Photodegradation assessment of RB5 dye by utilizing WO3/TiO2 nanocomposite: a cytotoxicity study. *Environ Sci Pollut Res* **29**, 22372–22390 (2022). https://doi.org/10.1007/s11356-021-17243-6
25. Su, C. *et al.* Effect of cephalexin after heterogeneous Fenton-like pretreatment on the performance of anaerobic granular sludge and activated sludge. *Chemosphere* **235**, 84–95 (2019). https://doi.org/10.1016/j.chemosphere.2019.06.136
26. Commercial Ministry of Islamic Republic of Iran, The Department of Statistic, Tariff, and goods report. https://www.amar.org.ir/english (2011).
27. Greenberg, A. E., Eaton, A. D. & Franson, M. A. H. *Standard methods for the examination of water and wastewater*. (APHA-AWWA-WEF, 2005).

28. Ghodsian, M. Removal of dye by the stabilized TiO2 nano-particles on concrete under UV irradiation. (Tarbiat Modares University).
29. Mahvi, A. H., Ghanbarian, M., Nasseri, S. & Khairi, A. Mineralization and discoloration of textile wastewater by TiO2 nanoparticles. *Desalination* **239**, 309–316 (2009). https://doi.org/10.1016/j.desal.2008.04.002
30. Saravanan, M., Sambhamurthy, N. P. & Sivarajan, M. Treatment of acid blue 113 dye solution using iron electrocoagulation. *CLEAN – Soil, Air, Water* **38**, 565–571 (2010). https://doi.org/10.1002/clen.200900278
31. Abo-Farha, S. A. Photocatalytic Degradation of Monoazo and Diazo Dyes in Wastewater on Nanometer-Sized TiO2. 13.
32. Pirkarami, A. & Olya, M. E. Removal of dye from industrial wastewater with an emphasis on improving economic efficiency and degradation mechanism. *Journal of Saudi Chemical Society* **21**, S179–S186 (2017). https://doi.org/10.1016/j.jscs.2013.12.008
33. Godini, K., Azarian, G., Rahmani, A. R. & Zolghadrnasab, H. Treatment of waste sludge: a comparison between anodic oxidation and electro-Fenton processes. *J Res Health Sci* **13**, 188–193 (2013).
34. Taniguchi, M. & Lindsey, J. S. Database of absorption and fluorescence spectra of >300 common compounds for use in photochem CAD. *Photochemistry and Photobiology* **94**, 290–327 (2018). https://doi.org/10.1111/php.12860
35. Damodar, R. A. & You, S.-J. Performance of an integrated membrane photocatalytic reactor for the removal of Reactive Black 5. *Separation and Purification Technology* **71**, 44–49 (2010). https://doi.org/10.1016/j.seppur.2009.10.025
36. Irani, M., Mohammadi, T. & Mohebbi, S. Photocatalytic degradation of methylene blue with ZnO nanoparticles; a joint experimental and theoretical study. *Journal of the Mexican Chemical Society* **60**, 218–225 (2016). https://doi.org/10.29356/jmcs.v60i4.115
37. Abtahi, S. M., Torabian, A., Vosoogh, A., Jafari, B. & Gholizadeh, M. Comparison of the monod and kincannon-stover models for kinetic evaluation in an anaerobic baffled reactor (ABR). *Environmental Sciences* **8**, 55–66 (2011).
38. Choi, H.-J. & Yu, S.-W. Biosorption of methylene blue from aqueous solution by agricultural bioadsorbent corncob. *Environmental Engineering Research* **24**, 99–106 (2019). https://doi.org/10.4491/eer.2018.107

Author contributions

All authors contributed to the study's conception and design. Material preparation, data collection and analysis were performed by [**Roya Nayebi Gavgani**], [**Bita Ayati**] and [**Hossein Ganjidoust**]. The first draft of the manuscript was written by [**Bita Ayati**] and all authors commented on previous versions of the manuscript. All authors read and approved the final manuscript.