

The influence of Operational Parameters on the Photocatalytic Decolorization of Tubantin blue BRR-HC dye in pure aqueous TiO2 Suspensions

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Abstract

Photo-catalytic decolorization of Tubantin blue BRR-HC dye was investigated using pure TiO_2 anatase in aqueous suspension under solar radiation. The influence of pH solution, amount of catalyst, dye concentration, volume of 30% hydrogen peroxide and irradiation time was she died. Temperature of solution and photon flux were calculated during the experiments. Results show that the employment of efficient photo-catalyst and selection of optimal operational parameters might lead to complete decolorization

المستخلص تتم التحقق من التحفيز الضو ي لأزاله لون صبغه التوبيتان الزرقاء باستخدام محلول مائي لثاني اوكسيد التيتانيوم النقي تحت أشعه الشمس وتمت دراسة تأثير الأس الهيدروج ي ،وكميه محفز ،وتركيز الصبغة ،وحجم محلول فوق اوكسيد الهيدروجين تركيزه 0% و زمن التعرض للأشعه علي التفاعل. تم حساب درجه حرارة المحلول وتدفق الفوتونات الثاء التجارب . وضحت النتائج أن تطبيق العوامل الفعالة ربما قادت لإزاله كامله للون الصبغه.

KEYWORD: Tubantin blue BRR-HC, Advance Oxidation processes titanium dioxide photocatalyst.

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is lost during the dyeing process and is

released into the environment as textile

effluent (Han et al.; 2009, Kerkez-

Kuyumcu et al., 2015). The release of those

colored was tewaters in the environment is

a considerable source of non-aesthetic

pollution and eutrophication and it can

originate dangerous byproducts through

oxidation, hydrolysis or other chemical

reactions taking place in the wastewater

phase (Habibi et al., 2005, Mahmood and

Arami, 2006, Kordouli et al., 2015).It

inhibits sunlight penetration into the stream

Introduction

Dyes are organic compounds that colorized materials. Dyes can be classified as anionic (direct, acid, and reactive dyes), cationic (basic dyes), non-ionic (disperse dyes), azo, diazo, anthroquinone based and metal complex dyes. (Mall et al., 2006). Large amounts of synthetic dyes are produced and used in many different industries; textiles, paper, cosmetic, printing, leather, rubber and plastics. (Seyyedi and Jahromi, 2014; Kerkez-Kuyumcu et al., 2015). About 1–20% of the total global production of dyes

and reduces the photosynthetic reaction. Some dyes are also toxic and even carcinogenic. (Shakir et al., 2010). Removing color from wastes is often more important than other colorless organic substances, because the presence of small amounts of dyes (over 1 ppm) is clearly and influences the visible water environment considerably (Verma et al., 2012). Highly substituted aromatic rings, joined by one or more azo groups, characterize their chemical structures. These substituted ring structures make the molecules recalcitrant which the conventional wastewater treatment, such as Biological physical, chemical and physico-chemical methods, which include coagulation and flocculation(Aleboveh et al.; 2008, Harrelkas et al., 2009; Zonoozi et al., 2009), membrane separation (Sachdeva and Kumar, 2009; Amini et al., 2011) sonochemical degradation (Abbasi and Asl, 2008), electrochemical (Gupta et al., 2007; Fan et al., 2008), activated carbon adsorption(Xu et al., 2008; Tan et al., 2008; Foo and Hameed, 2010) and adsorption (Crini, 2005, Crini and Badot, 2008) processes do not degrade (Soutsas et al., 2010; Ajma et al., 2016). These methods only succeed in transferring organic compounds from water to another phase, thus creating secondary pollution. This will require a further treatment of solid-wastes and regeneration of the adsorbent which will add more cost and expensive operations to the process(Mall et al., 2006;

Mittal et al., 2014; Lima et al., 2015). In recent years an alternative to conventional methods, that is "Advanced Oxidation Processes" (AOPs), which includes ozonation (Akpan and Hameed, 2009), H_2O_2/UV processes (Saquib et al., 2008)

Fenton and photo-Fenton catalytic reactions (Zangeneh et al., 2015) and heterogeneous photocatalysis based on the generation of very reactive species such as radicals hydroxyl that quickly and nonselectively, oxidizes a broad range of organic pollutants, was been applied. This technology leads to the total mineralization of the of most organic pollutants (Konstantinou and Albanis, 2004; Akpan and Hameed, 2009; Verma et al., 2012). nanophotocatalysis Heterogeneous has several advantages over competing processes are; complete mineralization, no waste-solids disposal problem and only mild temperature and pressure conditions are necessary(Seyyedi and Jahromi, 2014). TiO₂ semiconductor photo-catalyst, has received increasing attention because of its low cost, non-toxicity, relatively high chemical stability of the catalyst, and the possibility of using sunlight as a source of irradiation. The aim of the present work is to investigate the influence of various parameters photo-catalytic on decolorization of Tubantin dye (fig 1), in the presence of pure TiO_2 irradiated by the Solar light. And also to determine solution temperature and photon flux during the reactions.

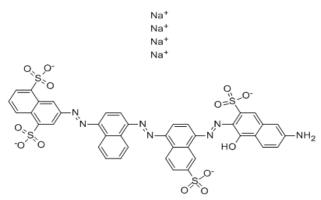


Fig (1): Chemical structure of Tubantin blue BRR-HC dye⁽³⁹⁾

Materials & Methods Materials Chemicals

Pure titanium dioxide (particle size 49nm, specific surface area 46.962 m²/g) from Aldrich , tubantin blue BRR-HC dye (colour index C.I DrB71) from Bezema, ferrioxalate actinometry (0.006M) was prepared, distilled water, sodium hydroxide 1%, sulphuric acid 0.5M, 1-10phenanthroline, Sodium acetate and all other chemicals of analytical grade.

Apparatus

Sensitive balance, magnetic stirrer, spectrophotometer (Jenway Aqunova), compound parabolic collector (CPC) solar collector (built by . Dr. Saleh Hamdo at Solar Energy Equipment Co), borosilicate tube (length 50cm,d 5cm, with slot on end to outlet the product.) built by technician Ahmed at physics Department – Faculty of science – Khartoum University

Method

Photo-reactor setup

All experiments were performed under the air and natural solar radiation. The solar CPC. was constituted by one CPC unit $(0.2m^2)$ and tilted 15° N local latitude. The cvlindrical photochemical reactor 800cm3capacity was made-up of borosilicate glass (length 50cm,d 5cm) (fig The slot 2). was opened during experimentation.



Fig (2):1 Photo-reactor setup

Determination of photon flux

2.947g of ferrioxalate actinometry (0.006M) solution was prepared in 100cm^3 of H₂SO₄(0.5 M) and diluted with distilled water to 1 dm³. 450 cm³ (V₁) of solution irradiated under solar light using photoreactor in (fig 2) for 30 min. 10cm^3 (V₂) of irradiated solution was transfered into 50

cm³ volumetric flask (V₃) containing a mixture of 5cm³ of 0.1% 1.10phenanthroline solution and 2.5 cm³ buffer pH 5, and then diluted to the mark with distilled water. A reference is prepared in the same way except that it has not been irradiated. Both solutions were kept in the dark (about 30 min) until full colour development is achieved, and the absorbance difference between the two samples were measured at 510nm [optical path length l=2.5 cm, ϵ (510nm) = 11100 dm3mol⁻¹cm⁻¹, Φ (510 nm) = 1.26 (0.006M)]. The photon flux amount $q_{n,p}$ einstein s-1was calculated by (Kuhn et al., 2004)

 $q_{n,p} = \frac{\Delta A V_1 V_3}{\Phi(\lambda) \varepsilon(510nm) V_2 lt} \quad -(1)$

Photodecolorization of dye

500 cm³ of dye solution was taken for all experiment, desired initial concentration for the different experiment (12.5, 25, 50, 100 and 200ppm) were prepared from a stock solution of 1000ppm. The pH was adjusted in (1.7, 3.3, 7.4, 9.4 and 10.6) using H₂SO₄ 1% or NH₄OH 1%. Desired amount of

catalyst (0.1, 0.125, 0.25, 1, 2 and 3g). Repeat the experiments using H₂O₂ (30%) in different volumes (12.5, 20, 25, 37.5, 50 cm³). In all experiments the reactor was washed with distilled water. Then the mixture was stirred for 30min. and exposed to solar radiation for two hours. The effect of irradiation time (1-6 hours) under optimum parameters. Then the absorbance was read at 436,525 and 620nm and the average were taken and were studied the percentage removal colour was determined using equation (2):

decolorizatin % =
$$\frac{A_i}{A_f} \times 100$$
 - (2)

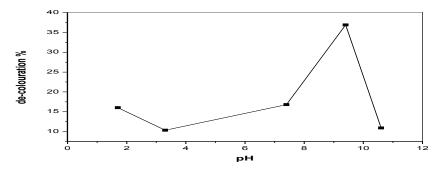
Where A_i is initial absorbance and A_f is final absorbance. The decolorization percentage versus irradiation time records using origin 8.6 computer programmer

Result and Discussion

Influence of pH

Table (1): Effect of pH on decolorization of tubantin blue dye at 50ppm concentration and 1g of catalyst

pН		Absorbance at 436nm	Absorbance at 525nm	Absorbance at 620nm	Average	De-coloration %
1.7	before	0.911	2.183	2.331	1.8083	16.03
	after	1.501	1.946	1.090	1.5183	
3.3	before	0.563	1.486	1.812	1.1930	10.34
	after	1.174	1.308	0.846	1.4512	
7.4	before	0.662	1.823	2.264	1.5830	16.80
	after	0.984	1.649	1.889	1.5123	
9.4	before	0.550	1.564	2.086	1.4000	36.90
	after	0.775	1.046	0.829	0.8833	
10.6	before	0.441	1.223	1.735	1.1330	10.94
	after	0.999	1.168	1.131	1.0090	



Fig(3) : Effect of pH on decoloration of Tubantin blue dye at 50ppm concentration and 1g of catalyst

The pH of the solution that achieved maximum removal was very effective, indicating that changes in pH of solution had а significant impact on the decolorization process. Table1 summarizes the influence of pH solution in photodecolorization and Fig (3) demonstrates the photo-catalytic decolorization of tubantin blue BRR-HC. The best results of Tubantin blue BRR-HC were obtained in alkaline pH 9.4, owing its molecular weight and structure cauing hindrance. Very high pH

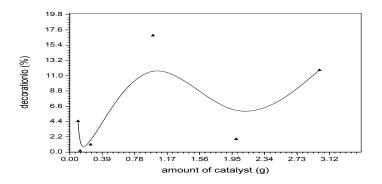
value was found to be favorable even when anionic azo dyes should hamper adsorption on the negatively charged surface. In alkaline solution OH radicals catalytic enhancement are easier to be generated by oxidation as more hydroxide ions are available on TiO_2 surface, thus the efficiency of the process is logically enhanced. (Habibi, al., 2005; et Konstantinou and Albanis, 2004; Akpan and Hameed, 2009; Qamar et al., 2005; Saquib et al., 2008)

Influence of Amount of Catalyst

Table (2): effect of amout o catalyst on decolorization of Tubantin blue dye at pH 7.4 dye at 50ppm concentration

Amount of catalyst g/dm ³		Absorbance at 436nm	Absorbance at 525nm	Absorbance at 620nm	Average	Decoloration %
0.1	before	0.662	1.823	2.264	1.58300	4.466
	after	0.928	1.720	1.889	1.51230	
0.125	before	0.675	1.798	2.242	1.57167	0.191
	after	0.996	1.786	1.924	1.56867	
0.25	before	1.046	2.480	2.500	2.00900	1.082
	after	1.304	2.041	2.249	1.74700	
1	before	0.662	1.823	2.264	1.583	16.8
	after	0.984	1.649	1.318	1.317	
2	before	0.521	1.552	2.120	1.4177	1.88
	after	1.341	1.693	1.342	1.391	
3	before	0.521	1.552	2.120	1.4177	11.8
	after	1.104	1.421	1.224	1.2497	

Table 2 demonstrates the effect of amount of catalyst on the photo-decolorization of Tubantin blue BRR-HC. Fig (4) show the effect of amount of TiO_2 on photodecolorization of Tubantin blue BRR-HC. Decolorization was observed to increase the photocatalyst amount.(Akpan and Hameed, 2009; Seyyedi and Jahromi, 2014; Sun et al., 2008) The increase in the amount of catalyst increased the number of active sites, on the photocatalyst surface, that in turn increase the number of hydroxyl and superoxide radicals causing an increasing the percentage of decolorization. (Huang et al., 2008). On the other hand when the concentration of TiO2 was decreased, the catalyst surface andits effect on absorption of light were limited.

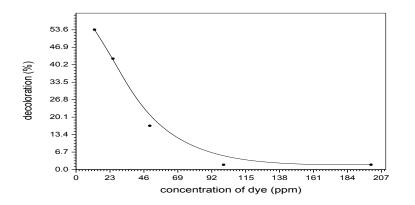


Fig(4) : effect of amount of catalyst(1/dm³TiO₂ pure) on decoloration of Tubantin blue dye at 50ppm concentration at pH 7.4

Influence of Initial Concentration of Dyes

Table (3) : concentration of dye on decoloration of tubantin blue dye at pH 7.4, catalyst (1g/dm³ TiO₂ pure)

Concentration of dye ppm		Absorbance at 436nm	Absorbance at 525nm	Absorbance at 620nm	Average	Decoloration %
12.5	before	0.176	0.425	0.635	0.412	53.46
	after	0.222	0.180	0.170	0.191	
25	before	0.343	0.825	1.226	0.807	42,54
	after	0.505	0.510	0.376	0.463	
50	before	0.662	1.823	2.264	1.583	16.80
	after	0.984	1.649	1.318	1.317	
100	before	1.070	2.406	2.485	1.987	1.857
	after	1.352	2.031	2.190	1.858	
200	before	1.046	2.480	2.500	2.009	1.875
	after	1.304	2.041	2.249	1.875	



Fig(5) : Concentration of dye on decoloration of tubantin blue dye at pH 7.4, catalyst (1g/dm³

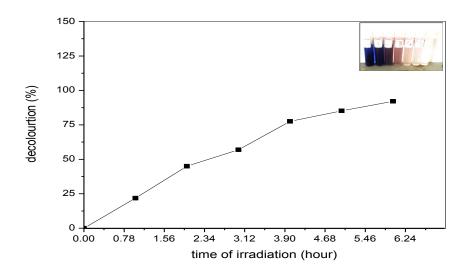
TiO₂ pure)

The effect of initial concentration of dye on photo-decolorization of dye was summarized in Table (3). Fig (5) shows that percentage declorization of tubantin blue BRR-HC decreases as the initial concentration of the dye increased. The decolorization was related not only to the probability of OH' radicals formation on the catalyst surface b tout also to the probability their reaction with dye molecules. It was postulated that when the initial concentration of dye is increased, more and more dye molecules are adsorbed on the surface of TiO2. whereas less number of photons are available to reach the catalyst surface and therefore less OH' radicals are formed.(Konstantinou and Albanis, 2004; Daneshvar et al., 2003) Influence of Time of Irradiation:

It can be seen from Table 5 and fig 6 percentage declorization increased as function of irradiation time . due to hydroxyl radical(OH[•]) has short half life time.

Table (4): effect of irradiation time on decoloration of Tubantin blue dye at 50 ppm at pH 9.4	
catalyst (1g/dm ³) under air	

Time of	Absorbance at	Absorbance at	Absorbance at	Average	Decoloration
irradiation	436nm	525nm	620nm		%
hour					
0	0.515	1.394	1.929	1.2793	0
1	0.730	1.148	1.124	1.00067	21.80
2	0.675	0.790	0.647	0.70400	45.00
3	0.679	0.567	0.402	0.54930	54.70
4	0.386	0.299	0.177	0.28730	77.50
5	0.272	0.179	0.116	0.15037	88.20
6	0.132	0.125	0.048	0.10167	92.01



Fig(6) : Effect of irradiation time on decoloration of tubantin blue dye at 50pmm, pH 9.4, catalyst (1g/dm³) under air

Temperature of solution:

Table (5) and fig (7) show the increase of temperature of solution during the decolorization of Tubantin blue BRR-HC. It was noticed that temperature was increased during experiment, owing the use

of CPC collector. Also related to daily period. However temperature is increased the decolorization uninfluenced. The best temperature range for the photo degradation is $20-80 \circ C$. (Carp et al., 2004)

Table(5): The changing of temperature of solution against the irradiation time during decolorization of Tubantin blue dye at 50ppm, pH 7.4, catalyst (1g/dm³) under air

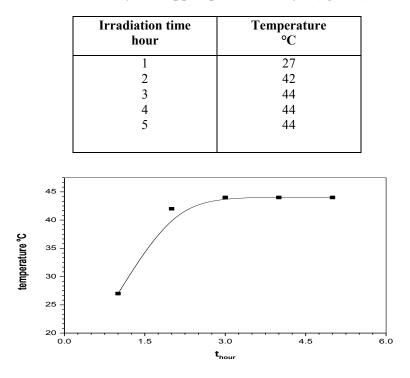


Fig (7): the various of temperature against the irradiation time during decolorization of Tubantin blue dye at pH 7.4, catalyst (1g/dm³) under air

variation in the values of photo flux Table((6) and Figure (8)) was quite consistent with the phenomenon of the daily change in solar light intensity.

Photon Flux

Solar light irradiation changed daily and even on the same day(Neppolian et al., 2002, Carp et al., 2004). Therefore, the

Table (6):photon flux during decolorization of tubantin blue dye at 50ppm, pH 9.4, catalyst (1g/dm³) under air

Irradiation time	Photon flux
hour	Enstien / second
First	7.615630E ⁻⁵
Second	5.212596E ⁻⁵
Third	3.643923E ⁻⁵
Fourth	2.494090E ⁻⁵
Fifth	3.671580E ⁻⁵

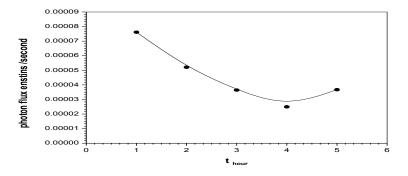


Fig (7): photon flux during decolorization of Tubantin blue dye at 50ppm, pH 9.4, catalyst (1g/dm³) under air

Influence of hydrogen peroxide H₂O₂

agents (hydrogen peroxide Oxidizing (H_2O_2) , ammonium persulphate bromate $((NH_4)_2S_2O_8)$ and potassium (KBrO₃) and oxygen) have a great deal of influence on the photo-catalytic degradation of dyes. (Shakir et al., 2010; Seyyedi and Jahromi, 2014; Saquib et al., 2008; Saquib et al., 2008). The effect of volume of 30% H₂O₂ on decolorization of tubantin blue

BRR-HC was performed by varying the volume of 30% H₂O₂ (12.5, 20, 25, 37.5 and 50 cm³). It can be seen from Table (7) and fig (9) the percentage of decolorization increased as volume of 30% H₂O₂ increased. This increase due to the effect of the additionally produced hydroxyl radicals. Fig (10) shows that TiO₂/H₂O2 system gave high decolorization percentage.

Table (7):effect of volume of $H_2O_2(30\%)$ on decolorization of tubantin blue dye at 50ppm, pH 9.4, catalyst (1g/dm³)

Concentration of dye ppm		Absorbance at 436nm	Absorbance at525nm	Absorbance at 620nm	Average	Decolorization %
12.5	before	0.560	1.992	2.008	1.5200	55.9211
	after	0.528	0.809	0.673	0.6730	
20	before	0.509	1.439	1.222	0.9923	34.5063
	after	0.6210	0.935	0.845	0.8003	
25	before	0.560	1.992	2.008	1.5200	84.4079
	after	0.270	0.289	0.152	0.2370	
37.5	before	0.727	1.238	1.302	1.0890	52.7090
	after	0.662	0.505	0.378	0.5150	
50	before	0.560	1.992	2.008	1.5233	91.35630
	after	0.179	0.130	0.086	0.1317	

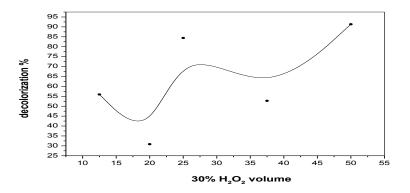


Fig (9):effect of volume of $H_2O_2(30\%)$ on decolorization of tubantin blue dye at 50ppm, pH 9.4, catalyst (1g/dm³)

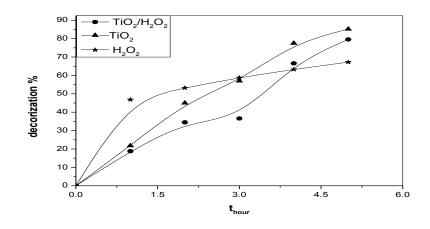


Fig (10): Decolorization % of Tubantin blue dye depending on H_2O_2/TiO_2

Conclusion

The progress of photo-catalytic decolorizeation of Tubantin blue dye during changing the experimental parameters was monitored by measuring the absorbance in uv-visible spectrum 436, 525 and 620nm. Results indicate that the photo-catalytic decolorizetion of tubantin blue dye in water by TiO₂ catalyst were diverse and determined by the concentration of the dye, pH of the solution, amount of photo-catalyst, volume of 30% hydrogen peroxide and irradiation time. During these investigations the temperature of the solution had increased as the photon flux are vary during the experiment.

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