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Investigation on basic blue 41 dye degradation by fenton reaction

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Abstract. Basic blue 41 is a very stable dye using in wool weaving industry. Fenton reaction is often used to decompose stable substances in wastewater. In this study planed experiments method was used to investigate the effect of three factors, that are pH, H_2O_2 and Fe^{2+} concentration on COD reduction. The response surface was determined by program Modde 5.0, the optimal reaction conditions was: Fe^{2+} concentration is 120 mg/L, H_2O_2 concentration is 10 mM, pH is 4.

1. Introduction

Dyes and pigments released into the environment mainly in the form of wastewater effluents by textile, leather and printing industries cause severe ecological problems. These compounds have a great variety of colors and chemical structures and are recalcitrant to microbial attack. Most of the dyes are non-toxic, except for azo-dyes which comprise a large percentage of synthetic dyes and are degraded into potentially carcinogenic amines [1-3]. Most of the dyes are non-toxic, except for azo-dyes which comprise a large percentage of synthetic dyes and are degraded into potentially carcinogenic amines [2].

Textilewastewaters offer considerable resistance to biodegradation due to presence of the dyestuffs which have a complex chemical structure and are resistant to light, heat and oxidation agents. Biological treatment processes such as aerated lagoons and conventional activated sludge processes are frequently used to treat textile effluents. These processes are efficient in the removal of suspended solids but largely ineffective in removing dyes from wastewater [3]. Chemical treatment systems, on the other hand, are generally more effective with respect to biological processes in decolorization of textile dyestuffs although their application is limited with their high costs [2-5].

The combination of hydrogen peroxide and a ferrous salt has been referred to as "Fenton's reagent". The primary oxidant in Fenton's reagent is the hydroxyl radical (•OH) generated by the reaction of hydrogen peroxide with ferrous ion [6]. The hydroxyl radical is very active and can react unselectively with the compounds in the reaction mixture including hydrogen peroxide and ferrous salt. Therefore

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ferrous salt and hydrogen peroxide concentrations have great influence on the effiency of Fenton reaction [7]. In this paper we used planned experiments to find out the optimum condition for Fenton reaction.

2. Material and methods

Wastewater containing basic blue 41 dye was purchased from the company "Det len Mua dong".

$FeSO_4.7H_2O$, H_2O_2 are pure chemicals (analytical grade).

Experimental procedure

200 ml textile wastewater was spilled to a glass, pH was adjusted by H_2SO_4 98%, Fe^{2+}, H_2O_2 were added at studying concentration, the mixture was stirred during 180 min. Afterthat the pH was adjusted to the value of 11-12, anticipated Fe^{3+} was removed by filtration. The solution was boiled under reflux to get rid of residual H_2O_2. COD of the wastewater was analysed according to the standard methods [8].

The factors influent on the Fenton reaction was investigated by planned experiments method.

COD removal efficiency depends on three factors: Fe^{2+} concentration, H_2O_2 concentration vµ pH. We have chosen the ranges to investigate as follows: pH = Z_1 = 2 – 6; H_2O_2 concentration: Z_2 = 5 – 15 mM; Fe^{2+} concentration: Z_3 = 50 – 150 mg/l.

The statistic model illustrating COD removal efficiency was as follows:

$$\hat{y} = b_o + b_1x_1 + b_2x_2 + b_3x_3 + b_{12}x_1x_2 + b_{13}x_1x_3 +$$
$$+ b_{23}x_2x_3 + b_{11}x_1^2 + b_{22}x_2^2 + b_{33}x_3^2 + b_{123}x_1x_2x_3$$

$\hat{y}$: response surface, COD removal efficiency (%).

x_1: coded variable of pH; x_2: coded variable of H_2O_2 concentration; x_3: coded variable of Fe^{2+} concentration;

3. Results and discussion

In this experiments we investigated the influence of three factors on the COD removal efficiency of wastewater: pH, H_2O_2 concentration, Fe^{2+} concentration. According to the previous research the investigating range was chosen: pH from 2 to 6, H_2O_2 concentration from 5 to 15 mM, Fe^{2+} concentration from 50 - 150 mg/l. The experiments were carried out as plan in the table 1. The fitted parameters and student errors are in Table 2. The parameter of term pH*[Fe^{2+}] is invalid so it was removed. The model validity R^2 and reproducibility Q^2 are close to 1 so we can conclude the model is fitted well.

Table 1. The COD reduction achieved from the planned experiments

Number of exp.	pH	H_2O_2 conc. (mM)	Fe^{2+} conc. (mg/l)	COD reduction (%)
1	2	5	50	56.3
2	6	5	50	50.4
3	2	15	50	55.1
4	6	15	50	47.2
5	2	5	150	68.2
6	6	5	150	64.2
7	2	15	150	65
8	6	15	150	55.8
9	0.636	10	100	61

10	7.364	10	100	48.6
11	4	1.59	100	64.5
12	4	18.41	100	59.2
13	4	10	15.9	52.35
14	4	10	184.1	68.2
15	4	10	100	79
16	4	10	100	76.9
17	4	10	100	77.8

Table 2. Regression coefficients and response

COD reduction	Coeff. SC	Std. Err.	P	Conf. int(±)
Constant	77.9036	0.560401	2.62951e-013	1.32516
pH	-3.50388	0.263151	3.15525e-006	0.622262
$[H_2O_2]$	-1.82414	0.263151	0.000224801	0.622262
$[Fe^{2+}]$	5.18805	0.263151	2.15807e-007	0.622262
pH*pH	-8.1777	0.289605	1.79551e-008	0.684816
$[H_2O_2]*[H_2O_2]$	-5.68577	0.289605	2.22113e-007	0.684816
$[Fe^{2+}]*[Fe^{2+}]$	-6.24248	0.289605	1.16604e-007	0.684816
$pH*[H_2O_2]$	-0.899997	0.343842	0.0345357	0.813066
$pH*[Fe^{2+}]$	0.0749973	0.343841	0.833561	0.813066
$[H_2O_2]*[Fe^{2+}]$	-0.899998	0.343842	0.0345356	0.813066
$y=77.90 - 3.50x_1 - 1.82x_2 + 5.18x_3 - 8.18x_1^2 - 5.69x_2^2 - 6.24x_3^2 - 0.90x_1x_2 – 0.90x_2x_3$				
N = 17	Q^2 = 0.975		Cond. no. = 4.9932	
DF = 7	R^2 = 0.996		Y-miss = 0	
Conf. level = 0.95	R^2 Adj. = 0.991		RSD = 0.9725	

Table 3. Analysis of variance

COD reduction	DF	SS	MS (variance)	F	p	SD
Total	17	66432.3	3907.78			
Constant	1	64822.1	64822.1			
Total Corrected	16	1610.22	100.639			10.0319
Regression	9	1603.6	178.178	188.385	0.000	13.3483
Residual	7	6.62071	0.945816			0.972531
Lack of Fit (Model Error)	5	4.40072	0.880143	0.792923	0.640	0.93816
Pure Error (Replicate Error)	2	2.22	1.11			1.05356
N = 17	Q^2 =0.975		Cond. no. = 4.9932			
DF = 7	R^2 = 0.996		Y-miss = 0			
	R^2 Adj. = 0.991		RSD = 0.9725			

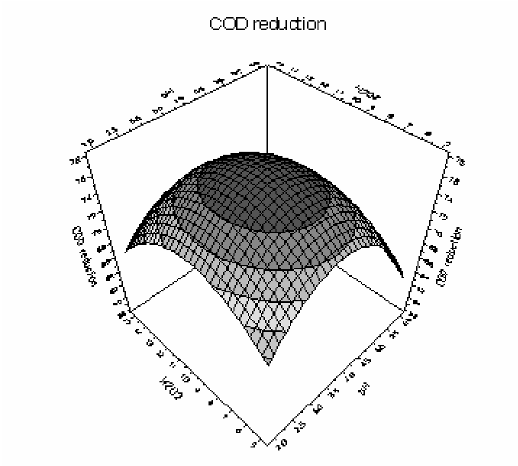

Fig. 1. Dependence of COD reduction on pH and H_2O_2 concentration when Fe^{2+} concentration remains constant.

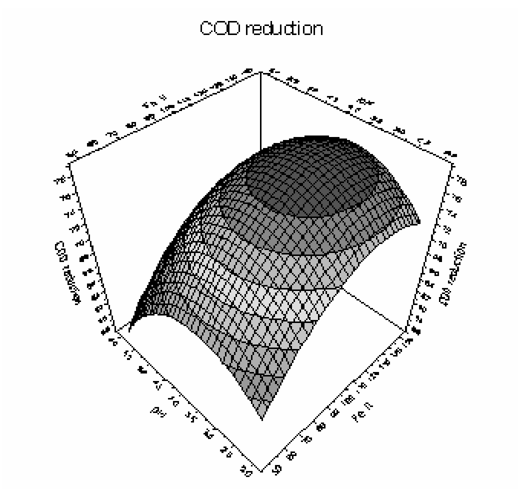

Fig. 2. Dependence of COD reduction on pH and Fe^{2+} concentration when H_2O_2 concentration remains constant.

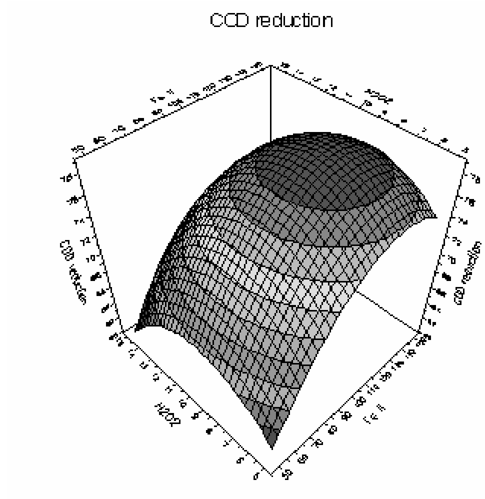

Fig. 3. Dependence of COD reduction on Fe^{2+} concentration and H_2O_2 concentration when pH remains constant.

The response illustrated dependence of COD reduction efficiency on pH, H_2O_2 and Fe^{2+} concentration can be formulated as follows:

$$y=77.90 - 3.50x_1 - 1.82x_2 + 5.18x_3 - 8.18x_1^2 - 5.69x_2^2 - 6.24x_3^2 - 0.90x_1x_2 - 0.90x_2x_3$$

According to the response and the fig. 1 – 3 we can notice that Fe^{2+} concentration had greatest influence on COD reduction efficiency, meanwhile H_2O_2 concentration had smallest influence. From the figures we can also deduce the optimal zone, the calculated results are listed in the table 4.

Table 4. Optimal reaction conditions

pH	H_2O_2 conc.(mM)	Fe (II) conc. (mg/l)	COD reduction
3.6554	8.9843	120.215	79.5149
3.6326	8.9826	120.843	79.522
3.6532	8.9693	120.47	79.5159
3.6407	8.9901	120.359	79.5191
4	10	120	78.9801
4	10	120	78.9801
4	10	120	78.9801
4	10	120	78.9801

Therefore we chose the optimal conditions for further studies as follows: Fe^{2+} concentration of 120 mg/L, H_2O_2 concentration of 10 mM, pH of 4.

Conclusion

The planned experiments allowed us to reduce the number of experiments and find out the optimal condition for the Fenton reaction which is used to degrade the basic blue 41 dye.

References

[1] K. Santhy, P. Selvapathy, Removal of reactive dyes from wastewater by adsorption on coir pith activated carbon, *Bioresource Technology* 97 (2006) 1329.

[2] U. Bali, B. Karagozoglu, Performance comparison of Fenton process, ferric coagulation and H_2O_2/pyridine/Cu(II) system for decolorization of Remazol Turquoise Blue G-133, *Dyes and Pigments*, 74 (2007), Iss. 1, 73.

[3] B. Noroozi, G.A. Sorial, H. Bahrami, M. Arami, Equilibrium and kinetic adsorption study of a cationic dye by a natural adsorbent - Silkworm pupa, *Journal of Hazardous Materials* B139, 167–174.

[4] P. Nigam, G. Armour, I.M. Banat, D. Singh, Physical removal of textile dyes and solid state fermentation of dye-adsorbed agricultural residues, *Bioresour. Technol.* 72(2000) 219.

[5] M.M. Davila-Jimenez, - M.P. Elizalde-Gonzalez - A.A. Pelaez-Cid, Adsorption interaction between natural adsorbents and textile dyes in aqueous solution, *Colloids and Surfaces A: Physicochem. Eng. Aspects* 254 (2005) 107–114.

[6] TLP. Dantas, V.P. Mendonca, H.J. Jose, A.E. Rodrigues, R.F.P.M. Moreira, Treatment of textile wastewater by heterogeneous Fenton process using a new composite Fe_2O_3/carbon. *Chemical Engineering Journal*, 118 (2006) Iss. 1-2, 77.

[7] A. Duran, J.M. Monteagudo, M. Mohedano (2006): Neural networks simulation of photo - Fenton degradation of Reactive Blue 4. *Applied Catalysis B: Environmental* 65 (2006) 127.

[8] *Standard Methods for Examination of Water and Wastewater*, 1995, Washington, USA.

Nghiên cứu phân hủy phẩm nhuộm basic blue 41 bằng phản ứng Fentơn

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Basic blue 41 là một loại phẩm nhuộm cation rất bền, được sử dụng trong công nghiệp dệt len. Phản ứng Fentơn thường được sử dụng để phân huỷ các hợp chất bền trong nước thải. Trong công trình nghiên cứu này chúng tôi đã sử dụng phương pháp qui hoạch thực nghiệm để khảo sát ảnh hưởng của ba yếu tố là pH, nồng độ H_2O_2, nồng độ Fe^{2+} tới hiệu suất xử lý COD. Hàm mục tiêu đã được xác định bằng phương pháp hồi qui, điều kiện tối ưu của phản ứng xác định được bằng thực nghiệm là: nồng độ Fe^{2+} là 120 mg/L, nồng độ H_2O_2 là 10 mM, pH bằng 4.

Keywords: textile wastewater, Fenton reaction.