Cumhuriyet Üniversitesi Fen Fakültesi
Fen Bilimleri Dergisi (CFD), Cilt:36, No: 3 Özel Sayı (2015)
ISSN: 1300-1949

Cumhuriyet University Faculty of Science
Science Journal (CSJ), Vol. 36, No: 3 Special Issue (2015)
ISSN: 1300-1949

The removal of methylene blue by modified alumina using sodium dodecyl sulphate(SDS) from aqueous solutions

Fereshteh IRANİ[1,*], Ayoub PARCHEH BAF JADİD[1]

[1] *Chemistry Department, Ardabil Branch, Islamic Azad University, Ardabil, Iran*

Received: 01.02.2015; Accepted: 05.05.2015

Abstract. In this approach we explained removal of methylene blue(MB) from aqueous solution as one of the reactive dye stuffs that is useful in textile dyeing. In this research, we optimized the removal procedure of MB from water using modified alumina(Υ-Al_2O_3) by Sodium dodecyl sulphate (SDS). In this study a response surface method (RSM) was utilized as an optimization method for optimization of effective parameters of the experiment. Four factors namely weight of Al_2O_3, initial concentration of MB(C_0), weight of SDS, and time were considered as main factors in five levels. It is found that all of these factors but C_0 were significant at a certain confidence level ($P<0.05$). Then, an empirical model was developed for the removal of MB from aqueous solution by modified alumina. An ANOVA analysis revealed that the validity of the predicted model. Plots of response surface and counter map and interaction plot for the predicted model showed that there was interaction between the weight of Al_2O_3 with itself. Results showed that the effect of the weight of SDS was the most important factor between the factors. Also the weight of alumina had medium effect among the factors. Finally, the removal of MB from different water samples was successfully done.

Keywords: Methylene Blue , Removal, Modified alumina , Sodium dodecyl sulphate, Water samples.

1. INTRODUCTION

Water is a vital compound and it has remarkable and unique characteristics, so it is one of the most important compounds which constitutes the great majority of organisms and the environment[1]. The growth of world population and utilizing of scarce water resources on one hand, they have been contaminated and consumption of water in industrial and agricultural places together with human activities, has been caused the contamination of water sources and it has created the water cisis in the world[2]. Any changes in physical, chemical, which have and biological properties water, air, soil, and food which have adverse effects on human health, the environment , and other organisms are called contamination[3,4]. Dye are environmental contaminants the exposure of them to human societies[5] cause the introduction of toxic materials to them can be dangerous and cause chronic poisoning[6]. Methyl orange[7] and methylene blue are to dyes which are used world wide[8]. MB has wide application, which include paper dyeing, temporary hair colorant, dyeing cotton, and wools[9,10]. It can diseases in humans such as heart beat increase, vomiting, shock, cyanosis, jaundice, quadriplegia ,and tissue necrosis[11]. Many methods including physical, chemical, and biological have been used for removal of pollutants from waste water. Some of them are; filtration[12], sedimentation[13], chemical oxidation[14], electrochemical processes[15], ion exchange[16], and adsorption[17].

In 2011, Murat et al., investigated the removal of MB from cotton stalk. In this study, was used cotton stalk as an adsorbent for removal of MB from aqueous solution. The effect of initial dye concentration, pH, temperature, adsorbent dosage, and the adsorption thermodynamics of MB was studied. The removal of dye was done at 90 min [18].

*Corresponding author. *Email address*: *Irani_fereshteh@ymail.com*

Special Issue: The Second National Conference on Applied Research in Science and Technology

In 2010, Zendehdel et al., utilized a semi-inpenetrating polymer for MB from wastewater sludge. They found that maximum dye adsorption efficiency was 95%. Also they showed that the synthesized polymer was a good membrane for the removal of cationic dyes from aqueous solutions[19].

In 2001, kanan et al., investigated the kinetics and mechanism of removal of MB by various carbons. They found that the efficiency of removal of MB dye increased with decreasing the initial concentration of MB and sorbent particle size, and increasing the contact time. They utilized Langmuir and freundlich adsorption isotherms in that research. The kinetics of adsorption process was first order with regard to intra-particle diffusion rate. Finally they concluded that the capacity of their adsorbent was comparable with the commercial carbon[20].

In 2014, mahmmodi et al., studied on the removal of MB by iron oxide using ultrasonic degradation. The effect of hydrogen peroxide on the removal efficiency of MB was studied. The effect of contact time and initial dye concentration on removal efficiency were studied too. Finally, the performance of dye removal using adsorption and ultrasonic techniques were compared[21].

In 2012 reza ansari et al., studied the adsorption of MB from aqueous solutions using modified sawdust by SDS, and investigated the effects of important parameters such as, pH, initial concentration of dye, amount of sorbent, and time[22].

In 2003, dogan et al., used perlite for the removal of methylene blue by adsorption process. The obtained results showed that this process carried out very fast and it had physical characteristics. The effect of different parameters such as initial concentration of dye, pH, and the temperature of solution was investigated. The result showed that the amount of removal of dye was increased by increasing the initial concentration of dye, pH, and temperature. It also showed that adsorption process obeys from second order kinetics model[23].

The main aim of this work is the removal of MB by modified alumina with sodium dodecyl sulphate. In this research, the optimization of several experimental parameters was investigated which are as follows; pH, the weight of alumina, the weight of SDS, time, and dye concentration.

2. EXPERIMENTAL

2.1. Materials

Υ -Al_2O_3, Methylene blue, Sodium hydroxide, Hydrochloric acid, Nitric acid, Sodium dodacyl solphate. were supplied by Merck co. and Υ - $Al_2O_3$was purchased from Fluka.

2.2. Methods

A given volume of MB solution was poured in a beaker. Then SDS was added to the MB solution and its pH was adjusted at 1.7. Then the prepared solution was placed in a volumetric flask and was diluted until desired volume. The pH and absorbance of this solution were read before addition of a given weight of Υ -Al_2O_3, after addition of Υ -Al_2O_3 the solution was agitated for 6 min. Finally, the solution was centrifuged for 30 min and the absorbance of solution was measured by UV-Vis spectrometer[24]. All of the amounts for the mentioned parameters were from Table1.

Table1. Factors and their levels in CCD(central composite design).

Factor					
Factor levels	-2	-1	0	+1	+2
Weight of Al_2O_3	0.5	1	1.5	2	2.5
MB(ppm)	20	40	60	80	100
Weight of SDS	0.04	0.06	0.08	0.1	0.12
Time	2	4	6	8	10

The removed efficiency was calculated from equation(1):

$$\mathbf{R\%=100\times(C_0-C_t)/C_0} \quad (1)$$

C_0 represents the initial concentration of the solution and C_t represents the remained concentration of MB at the time of t.

2.3. Data analysis

In this work, in order to optimize the effective factors in the removal of MB , software of Minitab ver.16.2. was used.

3. RESULT AND DISCUSSION

In this experiment, we studied the removal of MB From aqueous solutions it depends on: pH , contact time, weight of alumina, weight of SDS, and concentration of MB.

A central composite design(CCD) through 30 runs utilized for determination the main effects, and interaction effects[25]. Table 2 shows the experimental conditions and results for the removal of MB using CCD containing 6 central points.

Table 2. Orders of experiments and the results of removal efficiency.

RunOrder	Al_2O_3	SDS	t	C_0	R%
1	+	+	-	-	99.84
2	0	0	0	0	97.26
3	+	+	+	+	98.11
4	-	-	-	-	86.95
5	0	0	0	0	99.30
6	-	+	-	+	96.06
7	-	+	+	-	97.55
8	+	-	+	-	97.61
9	+	-	-	+	88.98
10	-	-	+	+	91.36
11	0	+	0	0	96.25
12	0	0	0	0	97.26
13	0	0	0	0	89.41
14	0	0	0	0	94.83
15	0	0	0	+	98.82
16	0	0	0	-	97.16
17	+	0	0	0	99.15
18	0	-	0	0	90.88
19	0	0	-	0	99.29
20	-	0	0	0	83.18
21	-	+	+	+	97.91
22	+	-	+	+	94.75
23	-	+	-	-	94.47
24	-	-	+	-	97.54
25	-	-	-	-	99.08
26	0	0	0	0	97.69
27	+	+	+	-	99.42

28	-	+	-	+	99.34
29	0	0	0	0	97.21
30	-	-	-	+	88.70

Regression coefficients and variance analysis is shown in table 3.

This table shows that weight of Al_2O_3 and weight of SDS, time, and $(Al_2O_3)^2$ have significant effects of the removal of MB.

Table 3. Statistical parameters for CCD.

Term	Coef	SE Coef	T	P
Constant	64.994	5.4560	11.912	0.000
Al_2O_3	19.585	6.3747	3.072	0.005
SDS	100.979	28.6235	3.528	0.002
t	0.846	0.2862	2.954	0.007
$Al_2O_3 \times Al_2O_3$	-4.902	2.0904	-2.345	0.027

This conclusion is true because the P-value for all of them is smaller than 0.05. it is clear that the amount of SDS is the most critical factor for the overall optimization procedure. Suitable models can be obtained using regression analysis. The estimated coefficients for the proposed models are given in table 4. The suggested empirical equation for our procedure is:

$$\mathbf{R=64.994 + 19.585\ Al_2O_3 + 100\ SDS + 0.846\ t - 4.902(Al_2O_3)^2} \quad (2)$$

Table 4. Estimated regression coefficients for R% for all of Factors.

Term	Coef	SE Coef	T	P
Constant	97.2583	1.067	91.179	0.000
Al_2O_3	4.8775	1.067	4.573	0.000
SDS	4.0392	1.067	3.787	0.002
t	3.3825	1.067	3.171	0.006
C_0	-1.1608	1.067	-1.088	0.294
$Al_2O_3 \times Al_2O_3$	-5.2854	1.996	-2.649	0.018
SDS× SDS	-2.8854	1.996	-1.446	0.169
t×t	-2.1004	1.996	-1.053	0.309
$C_0 \times C_0$	1.5396	1.996	0.772	0.452
$Al_2O_3 \times$ SDS	-1.2875	2.613	-0.493	0.629
$Al_2O_3 \times t$	-3.8825	2.613	-1.486	0.158
$Al_2O_3 \times C_0$	-3.0725	2.613	-1.176	0.258
SDS×t	-3.5675	2.613	-1.365	0.192
SDS× C_0	4.3825	2.613	1.677	0.114

t× C_0	-0.6825	2.613	-0.261	0.797

S = 2.61279

R-Sq = 81.35%   R-Sq(adj) = 63.95%

ANOVA table (Table 5) indicated that this regression is significant at 95% confidence level. The lack of fit is not significant (P-value=0.055). R^2=81.35% reveals that 81.35% of the total variation in the removal efficiency is explained by regression.

Table 5. ANOVA table for the parameters.

Source	DF	Seq SS	Adj SS	Adj MS	F	P
Regression	14	446.771	446.771	31.9122	4.67	0.003
Linear	4	317.362	317.362	79.341	11.62	0.000
Al_2O_3	1	142.740	142.740	142.740	20.91	0.000
SDS	1	97.889	97.889	97.889	14.34	0.002
t	1	68.648	68.648	68.648	10.06	0.006
C_0	1	8.085	8.085	8.085	1.18	0.024
Square	4	70.838	70.838	17.710	2.59	0.079
Al_2O_3×Al_2O_3	1	43.242	47.890	47.890	7.02	0.018
SDS×SDS	1	14.091	14.273	14.273	2.09	0.169
t×t	1	9.422	7.563	7.563	1.11	0.309
C_0×C_0	1	4.063	4.063	4.063	0.60	0.452
Interaction	6	58.571	58.571	9.762	1.43	0.267
Al_2O_3×SDS	1	1.658	1.658	1.658	0.24	0.629
Al_2O_3×t	1	15.074	15.074	15.074	2.21	0.158
Al_2O_3×C_0	1	9.440	9.440	9.440	1.38	0.258
SDS ×t	1	12.727	12.727	12.727	1.86	0.192
SDS×C_0	1	19.206	19.206	19.206	2.81	0.114
t×C_0	1	0.466	0.466	0.466	0.07	0.797
Residual Error	15	102.400	102.400	6.827		
Lack-of-Fit	10	92.147	92.147	9.215	4.49	0.055
Pure Error	5	10.254	10.254	2.051		
Total	29	549.172				

By comparing the efficiency of the run in table2, it is obvious that maximum R% belongs to the following conditions; C_0=40 ppm , weight of SDS=0.1, weight of Al_2O_3=2g, t=6min and pH=2, R%=99.84.

3.1. Optimum conditions and response surface plots

By making use of MINITAB software, the optimum removal of MB was found at C_0=40ppm, weight of SDS=0.1g, weight of Al_2O=2g and t=6min. These results belong to 99.84% removal efficiency. Two-dimensional and three-dimensional plots of response surfaces can interpret the influence of various factors on the removal efficiency.

The effect of weight of Al_2O_3 and weight of SDS on the removal efficiency has been shown in figures 1. It is clear that the weight of 2g for alumina and 0.1g for SDS is in the region of 97.5%-100% of removal efficiency when the time of experiment is 6min. figure 1(a) and 1(b) show this conclusion in three-dimensional and two-dimensional plots.

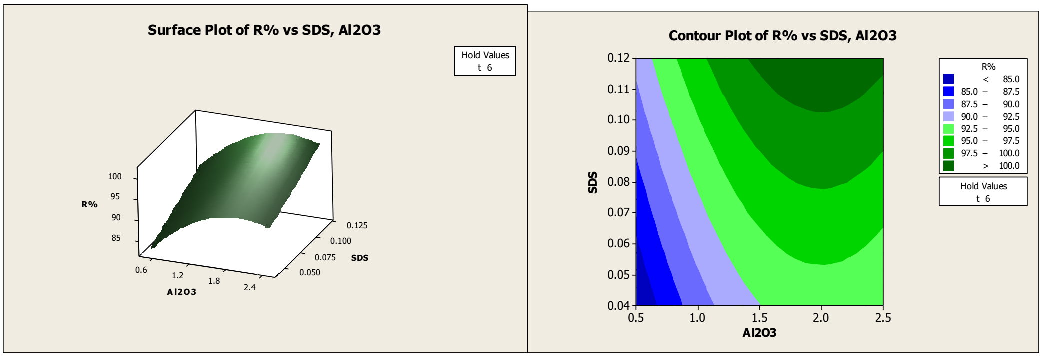


Figure 1(a) **Figure 1(b)**

Figure 1. Three-dimensional 1(a) and two-dimensional 1(b) plots of removal efficiency. Effective variables are Al_2O_3 and SDS, The middle level of time was chosen(6min).

Figure 2 shows the influence of Al_2O_3 and time on the removal efficiency. Figures of 2(a) and 2(b) indicate that time of 6 min, and 2g of alumina cause to 97.5-100% of removal efficiency when is 0.08g.

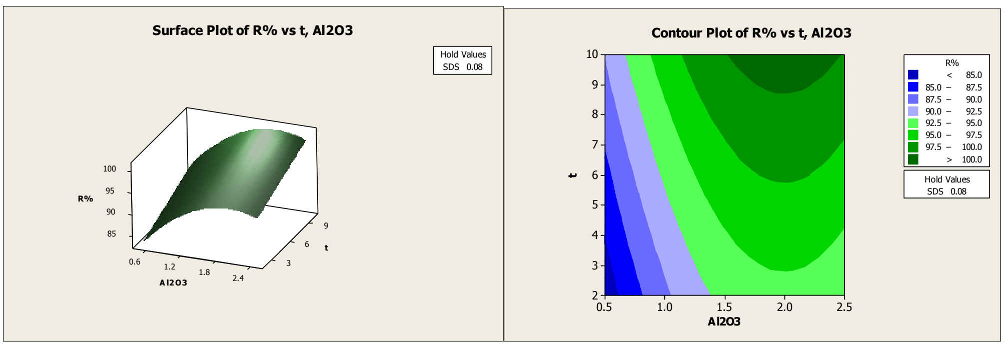
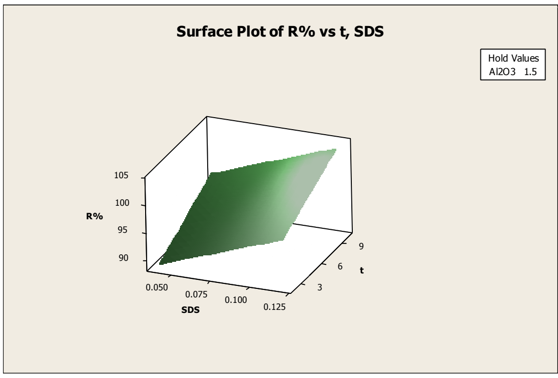


Figure 2(a) **Figure 2(b)**

Figure 2. Three-dimensional 2(a) and two-dimensional 2(b) plots of removal efficiency. Effective variables are Al_2O_3 and time, the middle level of weight of SDS was chosen(0.08g).

Figure 3 shows the influence of SDS and time on the removal efficiency. Figures of 3(a) and 3(b) indicate that time of 6min, and 0.08g of SDS cause to 100%-102% of removal efficiency when Al_2O_3 is 1.5g.



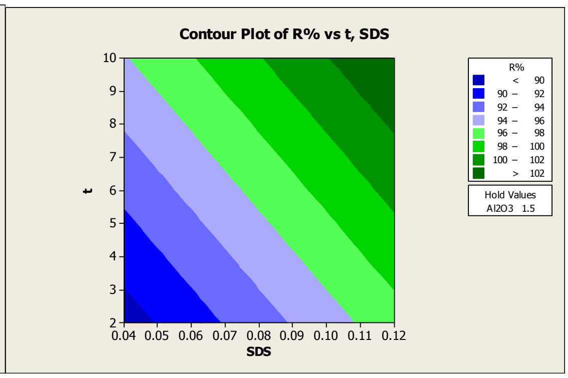


Figure 3(a) **Figure 3(b)**

Figure 3. Three-dimensional 3(a) and two-dimensional 3(b) plots of removal efficiency. Effective variables are SDS and time, the middle of weight of Al_2O_3 was chosen (1.5g).

4. CONCLUSIONS

In this research modified alumina was used as an excellent sorbent for the removal of MB from aqueous solutions. For factors namely; time, weight of alumina(A), weigh of SDS(S), and concentration of MB were chosen as effective parameters in this research. It is concluded that the concentration of MB no significant effect on the removal efficiency. So the most important factors were as follows; weight of alumina, weight of SDS, and time.

ACKNOWLEDGMENT

The authors would like to thank Islamic Azad university of Ardabil for the financial support of this research.

REFERENCES

[1] Helsel, D.R. and Hirsch, R.M. (2002), "Statistical Methods in Water Resources,"USGS Science for a Changing World, pp 1-524.
[2] Ratna. and Padhi, B.S. (2012),"Pollution Due to Synthetic Dyes Toxicity & Carcin Ogenicity Studies and Remediation," International Journal of Environmental Sciences, 3, pp 940-955.
[3] Qing lu, W. Hua Xie, S. Shan Zhow, W. Hui Zhang, Sh. Lin Liu, A. (2008),"Water Pollution and Health Impact in China," Open Environment Sciences, 2, pp 1-5.
[4] Singh Gambhir, R. Kappor, V. Nirola, A. Sohi, R. and Bansal,V. (2012),"Water Pollution : Impact of Pollutants and New Promising Techniques in Purification Process," Kamla-Raj, 37(2), pp103-109.
[5] Puvaneswari,N. Muthukrishnan, J. and Gunasekaran, P. (2006), "Toxicity Assessment and Microbial Degradation of Azo Dyes," Indian Journal of Experimental Biology , 44, pp 618 - 626.

[6] Kumar Samanta, A. and Agarwal, p. (2009), "Application of Natural Dyes on Textiles," Indian Journal of Fibre & Textile Research, 34, pp 384-399.
[7] Sathiyabama,J. Rajendran, S. Arockia Selvi, J. John Amalraj, A. (2008)," Methylene Orange As Corrosion Inhibitor for Carbon Steel in Well Water ," Indian Journal of Chemical Technology, 15, pp 462-466.
[8] Hashina, T. (2007), " The Rise of Japanese Sythetic Dye Industry During the First World War," Kobe University Economic Review, 53, pp 35-55.
[9] Miclescu, A . Wikluna, L. (2010), "Methyleme Blue, an Old Drug With New Indications? ," Jurnalul Roman De Anestezie Terapie Intensiva, 17 , pp35-41.
[10] Packianathan, N. and Karum Bayaram, S . (2010),"Formulation and Evaluation of Herbal Hair Dye : An Ecofriendly process," Journal of Pharmaceutical Sciences and Research, 2(10), pp 648-656.
[11] Tantiado, Rey G. (2012),"Comparative Mutagenic Effects of Laboratory Dyes on Drosphila Melanogaster," International Journal of Bio-Science and Bio-technology, 4 , pp 55-62.
[12] Koenigstein, N. Lanckriet,G. Mcfee, B. and Shavitt, Y. (2010), " Collaborative Filtering Based on p2p Networks," International Society For Music Information Retrieval Conference, pp153-158.
[13] Vogel, H. Wessels, M .Albrecht,C. Stich, H-B. and Wagner, B. (2010),"Spatial Variability of Recent Sedimentation in Lake Ohrid," Biogeosciences, 7 , pp3333-3342.
[14] Huang ,D. Wang, Y.J. Yang , L.M. and Luo, G.S. (2006), "Chemical Oxidation of Dibenzothiophene With a Directly Combined Amphiphilic Catalyst For Deep Desulfurization," Ind. Eng . Chem. Res. , 45 , pp1880-1885.
[15] Chandra Mouli, P. Venkata Mohan, S. and Jayarama Reddy, S. (2004), "Electrochemical Processes For the Remediation of Waste Water and Contaminated Soil: Emerging Technology ," Journal of Scientific & Industrial Research, 63, pp11-19.
[16] Hassig, M. Wullschleger, M. Naegeli,H . Kupper,J. Spiess, B. Kuster, N. Capstick, M. and Murbach, M. (2014), " Influence of Non ionizing Radiation of Base Stations on the Activity of Redox Proteins in Bovines," BMS Veterinary Research, 10 , pp1-11.
[17] Naujok, Roberta R. Paul, Hillary J. and Corn, Robert M. (1996),"Optical Second Harmonic Generation Studied of Azobenzene Surfactant Adsorption and Photochemistry at the water / 1,2-Dichloroethane Interface," The Journal of Physical Chemistry , 25, pp10497-10507.
[18] Ertas , M. Acemioglu, B. Hakki Alma, M. and usta ,M. (2011), "Removal of Methylene Blue From Aqueous Solution Using Cotton Stalk," Environmental Earth Sciences , pp899-906.
[19] Zendehdel,M . Barati,A. Alikhani,H. and Hekmat ,A. (2010), "Removal of Methylene Blue Dye From Waste Water by Adsorption Onto Semi-inpentrating Polymer Network Hydorogels Composed of Acryl amide and Acryl acid Copolymer and Polyvinyl Alcohol," Iran. J. Environ . Heath . Sci. Eng. , 7 , pp 423-428.
[20] Kanan, N. Meenakshi Sundaram, M. (2001), "Kinetics and Mechanism of Removal of Methylene Blue by Adsorption on Various Carbons – a comparative Study," Elsevier, 51 , pp25-40.
[21] Mahmoudi, Z. Azizian, S. Lorestani, B. (2014), " Removal of Methylene Blue from Aqueous Solution: A Comparison between Adsorption by Iron Oxide Nanospheres and Ultrasonic Degradation ," J.Mater . Environ. Sci. , 5(5) , pp 1332-1335.
[22] Ansari, R. Seyghali,B. (2012), "Application of Nano Surfactant Modified Biosorbent as an Efficient Adsorbent for Dye Removal," International Journal of Current Research , 47, pp 1802-1812.
[23] Dogan, M. Alkan, M. (2003), "Removal of Methyl Violent from Aqueous Solution by Perlite," J.Colloid Interface Sci, 267, pp 32-41.

[24] Zolgharnein, J. Adhami, Z. Shahmoradi, A. and Norollah Mousavi, S. (2010), "Optimization of Removal of Methylene Blue by Platanus Tree Leaves Using Response Surface Methodology," The Japan Society for Analytical Chemistry, 26, pp 111-116.

[25] Adlnasab, L. Ebrahimzadeh, H. Yamini, Y. and Mirzajani, F. (2010)," Optimization of a Novel Methods Baed on Solidification of Floating Organic Droplet by High-performance Liquid Chromatography for Evaluation of Antifungal Drugs in Biological Samples," Talanta, 83, pp370-378.