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Comparison and optimization of xylene cyanol uptake by natural and synthetic adsorbents

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ABSTRACT

The main objective of our work involves comparison of Xylene cyanol dye uptake among activated carbon and dried seaweeds (Green algae, Red algae). Equilibrium characteristics and behavior of three different adsorbents were investigated by performing batch adsorption experiments under static conditions by varying Xylene Cyanol concentration, pH, contact time, temperature and adsorbent dose were evaluated experimentally. Response Surface Methodology (RSM) based Box-Benkhen Design (BBD) was used to determine the effects of four influencing variables such as Dye concentration (mg/L), temperature (◦C), dose (g/L) and incubation time (hours) on Xylene Cyanol percentage removal for both activated carbon and dried seaweeds. The experimental results were found to be 82.08%, 73.86% and 53.76 for activated carbon, dried green algae and red algae respectively at their optimal conditions and found to be in agreement with predicted conditions.

Keywords: Xylene cyanol, adsorbent, optimization.

INTRODUCTION

Colored dye wastewater arises as a direct result of the release of the dye by textile industries (Allen, 2005). There are more than 100,000 commercially available dyes with over $7x10^5$ tones of dyes produced annually (Robinson, 2001). The release of dyes from textile industries in developing and under developing countries is a threat to ecosystem (Chen, 2003). Approximately 10-15% of the dyes discharged from dye production industries are major concern, as the colored effluents interferes with the penetration of light into the water, thereby leading to disturbances in the aquatic life (Ramakrishna, 1997; Karim, 2006). Non-biodegradable contaminants which are mostly removed by tertiary treatment pose a serious health and environmental hazard and they are not eco-friendly (Kannan, 2001). The drawback has made sorption technology as an alternative technique for uptake/removal of dyes due to its inherent low cost, simplicity, versatility and robustness (Volesky, 2001). Activated carbon is one of the most powerful adsorbent (Deva Prasath, 2011). In this study three different adsorbents Activated carbon, Green algae (*Chaetomorpha spiralis)*, Red and Brown algae (*Rhodophyceae and Phaephyceae)* were used.

Xylene cyanol is an acid dye used in the textile industry for dyeing of all natural fibers, e.g. wool, cotton, silk and synthetics, e.g.polyesters, acrylic and rayon. They are also used in a variety of application fields such as in paints, inks, plastics and leather. It is used a dye marker for agarose or acrylamide gel electrophoresis and is also suitable as a tracking dye for DNA sequencing. Its molecular mass is 538.61 g/mol, molecular formula is $C_{25}H_{27}N_2NaO_6S_2$ and its λmax is 612nm.

Response Surface Methods are designs and models for working with continuous treatments when the goal is to find the optima or to describe the response (Oehlert, 2000).It has also been successfully applied to different processes for achieving its optimization using experimental designs. In this work, we attempt to use locally available Activated carbon/Red and Brown algae as an adsorbent to remove/uptake Xylene cyanol 10 from aqueous solution and to investigate the interaction among physiochemical variables and to optimize the working conditions to predict optimal conditions.

MATERIALS

The organic dye Xylene cyanol was purchased from Himedia limited. All other chemicals used in the study were of analytical grade and procured from SD Fine Chemicals limited, India. Green algae (*Chaetomorpha spiralis)*, and Red marine seaweed residues (*Kappaphycus alwarezii)* were collected from marine coastal areas. All the adsorbents used were shade dried and powdered.

METHODOLOGY

Biosorption Experiment: Physico-chemical parameters such as pH, temperature(°C), effect of dye concentration (mg/L), incubation time (Hours), effect of adsorbent dose (g/L). The reaction mixture consists of 0.5g of adsorbent and 100mL of 50mg/L of standard working concentration of xylene cyanol, were incubated at room temperature under dark conditions without shaking. Samples were aliquoted at every two hours, filtered using whatmann filter paper and the filtrate was used for further analysis. The effect of pH was studied at varying pH (2, 3, 4, 5, 6, 7 and 8) at standard reaction conditions as stated above. The effect of temperature was studied at varying temperature

(30, 40, 50, 60 and 70 C) at optimum pH. The effect of dye concentration and incubation was studied at varying concentration from 50-250 µg/ml at its optimum pH and temperature for ten hours. Similarly the effect of adsorbent was studied at varying adsorbent concentration of 0.5g to 2.5g at optimum pH, temperature and dye concentration respectively. Initial concentration and final absorbance was recorded in UV spectrophotometer at 612nm and adsorption capacities of the adsorbent were calculated. The dye uptake capacity for each sample was calculated according to mass balance on the dye using Equation 01

$$qe = \frac{(ci - ce)V}{M} \quad \text{(Hong Zheng, 2009)} \quad \text{----------------- (01)}$$

Where C_i and C_e (mg/L) are the initial and final concentration of solution, respectively, V (L), the volume of Xylene cyanol solution used and W (g), the weight of the adsorbent used in the present study.

The sorption uptake percentage was calculated by using following equation:

$$sorption\,(\%) = \frac{(ci - ce)100}{ci} \quad \text{--------------- (2)}$$

RESULTS AND DISCUSSION

Adsorption studies: Xylene cyanol is an acid dye having toxic effects on living organisms, the effect of five influencing variables such as dye concentration, dose, pH, incubation time, temperature were investigated and optimized to predict the working range.

Effect of dye concentration on Dye removal: The effect of dye concentration was studied in the range of 50-250µg/mL. The adsorbate dosage is an important factor in sorption studies. Results indicate that increase in concentration simultaneously increases sorption uptake. The initial dye concentration acts as a driving force in overcoming resistance to the mass transfer of dye molecules from liquid phase and the solid phase, indicating that surface saturation is dependent on the dye concentration. The maximum adsorption capacity was achieved, as shown in the table for different adsorbents.

Effect of dose (Activated carbon/Green algae/Red marine seaweed residues) on Dye removal: The impact of adsorbate dose was investigated at a constant dye concentration (200µg/mL), indicating a linearity with sorption uptake. The sorption uptake increased linearly with incubation time and was maximum at an adsorbent dose of 1.5 g/mL at 10^{th}Hr of incubation. The maximum adsorption capacity was achieved, as shown in the table for different adsorbents.

Effect of pH on Dye removal: Adsorption of MB on silt increases with the increase of pH indicating favorable adsorption at alkaline medium. Sorption of Xylene cyanol on Sorption uptake efficiency is stable at acidic and at near neutral conditions, but decreases as increase in the pH value at alkaline conditions. Higher sorption might be due high hydrogen ion concentration, electron shuttling among the aromatic ring and ionization efficiency, leading to development of new adsorption sites. The maximum adsorption capacity was achieved, as shown in the table for different adsorbents.

Effect of temperature on Dye removal: Temperature is one of the influencing factors in a biosorption process. When temperature increase sorption uptake also increases in some point it going to be desorbed, that point removal of sorption decreased. The maximum adsorption capacity was achieved, as shown in the table for different adsorbents.

Effect of incubation time on Dye removal: Incubation time is also one of the influencing factors in a biosorption process. When time increase sorption uptake also increases in some point it going to be desorbed, that point removal of sorption decreased. The maximum adsorption capacity was achieved, as shown in the table for different adsorbents.

Table.1.Result of Adsorption studies

Adsorbent	Dye concentration	dose	pH	Temperature	Time
Activated carbon	200µg/mL	1.5g	5	50ºC	4 hours
Green Algae	200µg/mL	0.5g	4	70 ºC	6 hours
Red marine seaweed Residue	200µg/mL	0.5g	8	30 ºC	2 hours

Table.2.RSM for Activated carbon

	Factor 1	Factor 2	Factor 3	Factor 4	Actual response	Predicted response
Run	A:Dye concentration	B:Dose	C:Time	D:Temperature	R1	R2
	mg/L	g/100mL	Hours	◦C		
1	200	0.5	4	40	57.38	57.01
2	200	1	5	40	71.82	72.32
3	200	1.5	5	50	73.44	71.09
4	250	1.5	5	40	82.08	80.6
5	250	1	6	40	40.5	37.02
6	150	1	4	40	49.37	49.79
7	200	0.5	6	40	48.05	45.78
8	200	0.5	5	50	38.46	40.09
9	200	1	5	40	56.06	55.14
10	250	1	4	40	78.43	79.82
11	250	1	5	50	68.18	66.95
12	200	1	6	50	66.02	67.09
13	200	1.5	6	40	28.92	38.04
14	200	1	4	30	60	50.77
15	200	1	4	50	33.75	43.13
16	200	0.5	5	30	61.73	52.77
17	150	0.5	5	40	24.14	28.65
18	200	1	5	40	50	51.33
19	200	1	5	40	42.11	42.46
20	250	0.5	5	40	47.42	44.59
21	200	1	6	30	77.38	68.5
22	150	1.5	5	40	63.86	75.3
23	150	1	6	40	73.42	63.66
24	250	1	5	30	68.67	79.22
25	200	1	5	40	75.6	73.4
26	200	1.5	5	30	83.72	73.4
27	200	1.5	4	40	65.43	73.4
28	150	1	5	30	75.61	73.4
29	150	1	5	50	66.67	73.4

Table.3.RSM for Green Algae

	Factor 1	Factor 2	Factor 3	Factor 4	Actual response	Predicted response
Run	A:Dye concentration	B:Dose	C:Time	D:Temperature	R1	R2
	mg/L	g/100mL	Hours	◦C		
1	225	0.5	2	60	64.47	64.79
2	225	0.625	4	60	62.68	62.96
3	225	0.5	4	70	64.04	63.41
4	225	0.625	2	50	67.52	67.26
5	225	0.75	2	60	72.81	73.74
6	225	0.75	6	60	72.54	73.29
7	200	0.625	4	70	69.39	68.7
8	250	0.625	6	60	68.93	68.06
9	250	0.5	4	60	69.56	66.42
10	225	0.5	4	50	71.82	68.19
11	200	0.625	6	60	59.09	62.04
12	250	0.625	4	50	59.83	62.29
13	200	0.75	4	60	71.72	69.87
14	225	0.625	4	60	72.73	72.32
15	225	0.625	4	60	70.59	70.31
16	225	0.625	6	70	69.61	70.78
17	225	0.625	4	60	70.93	71.87
18	225	0.625	2	70	73.91	74.98
19	200	0.5	4	60	73.86	73.41
20	250	0.625	4	70	72.65	72.33
21	225	0.75	4	50	64.08	66.87
22	200	0.625	4	50	60.78	63.08
23	225	0.625	4	60	58.16	56.48
24	225	0.5	6	60	65.38	63.2
25	225	0.75	4	70	60.61	61.26
26	250	0.625	2	60	61.62	61.26
27	225	0.625	6	50	60.78	61.26
28	200	0.625	2	60	60.78	61.26
29	250	0.75	4	60	62.5	61.26

Table.4.RSM for Red and Brown Algae

	Factor 1	Factor 2	Factor 3	Factor 4	Actual response	Predicted response
Run	A:Dye concentration	B:Dose	C:Time	D:Temperature	R1	R2
	mg/L	g/100mL	Hours	◦C		
1	200	50	5	0.75	43.37	43.57
2	200	70	5	0.5	54.74	48.72
3	150	50	8	0.75	53.12	56.46
4	200	50	5	0.75	50.79	51.90
5	200	50	5	0.75	48.96	42.20
6	200	50	5	0.75	50.54	49.03
7	150	30	5	0.75	49.59	48.42
8	200	50	2	0.5	42.55	46.62
9	150	50	5	1	43.16	51.19
10	250	70	5	0.75	44.56	51.90
11	200	30	8	0.75	52.44	53.5
12	200	30	2	0.75	53.01	53.39
13	250	30	5	0.75	24.19	38.85
14	200	30	5	1	42.17	47.09
15	250	50	2	0.75	38.08	41.57
16	250	50	5	0.5	55.65	49.40
17	200	70	2	0.75	59.79	52.50
18	200	50	2	1	48.96	44.59
19	250	50	5	1	48.19	46.82
20	200	70	8	0.75	53.76	53.31
21	250	50	8	0.75	49.47	44.60
22	150	50	5	0.5	49.60	47.34
23	200	50	8	1	44.68	41.20
24	200	30	5	0.5	55.43	54.55
25	150	70	5	0.75	50.06	55.55
26	200	50	8	0.5	57.72	55.55
27	150	50	2	0.75	54.64	55.55
28	200	70	5	1	59.79	55.55
29	200	50	5	0.75	55.56	55.55

Table.5.ANOVA for Activated carbon

Source	Sum of Squares	Degree of freedom	Mean Square	F Value	p-value Prob> F
Model	6544.51	14	467.46	6.28	0.0007*
A-Dye concentration	462.02	1	462.02	6.20	0.0259
B-Dose	375.31	1	375.31	5.04	0.0414
C-Time	37.56	1	37.56	0.50	0.4891
D-Temperature	0.65	1	0.65	0.008	0.9267
AB	8.41	1	8.41	0.11	0.7417
AC	105.58	1	105.57	1.49	0.2534
AD	150.43	1	150.43	2.02	0.1770
BC	2.40	1	2.40	0.03	0.8600
BD	19.23	1	19.23	0.25	0.6192
CD	85.19	1	85.19	1.14	0.3028
A^2	92.89	1	92.89	1.25	0.2827
B^2	2.63	1	2.63	0.04	0.8535
C^2	5035.76	1	5035.76	67.66	< 0.0001
D^2	36.41	1	36.41	0.49	0.4957
Residual	1041.95	14	74.42		
Lack of Fit	816.91	10	81.6	1.45	0.3839
Pure Error	225.04	4	56.26		
Cor Total	7586.46	28			

Table.6.ANOVA for Green algae

Source	Sum of Squares	Degree of freedom	Mean Square	F Value	p-value Prob> F
Model	692.16	14	49.44	9.60	< 0.0001*
A-Dye concentration	3.06	1	3.06	0.59	0.4536
B-Dose	6.39	1	6.39	1.24	0.2839
C-Time	0.91	1	0.91	0.18	0.6801
D-Temperature	79.10	1	79.10	15.36	0.0015
AB	8.03	1	8.03	1.56	0.2321
AC	4.39	1	4.39	0.85	0.3716
AD	0.58	1	0.58	0.11	0.7427
BC	0.99	1	0.99	0.19	0.6678
BD	27.67	1	27.67	5.37	0.0361
CD	0.009	1	0.009	0.001	0.9672
A^2	52.23	1	52.23	10.14	0.0066
B^2	1.70	1	1.70	0.33	0.5742
C^2	531.42	1	531.42	103.18	< 0.0001
D^2	2.65	1	2.65	0.51	0.4852
Residual	72.10	14	5.15		
Lack of Fit	69.55	10	6.95	10.90	0.0171*
Pure Error	2.55	4	0.64		
Cor Total	764.26	28			

Table.7.ANOVA for Red and brown algae

Source	Sum of Squares	Degree of freedom	Mean Square	F Value	p-value Prob> F
Model	754.70	14	53.91	1.06	0.4563
A-Dye concentration	0.26	1	0.26	0.005	0.9445
B-Temp	193.84	1	193.84	3.82	0.0710
C-Time	19.03	1	19.03	0.37	0.5503
D-Dose	10.85	1	10.85	0.21	0.6510
AB	23.52	1	23.52	0.46	0.5072
AC	67.24	1	67.24	1.32	0.2691
AD	0.17	1	0.17	0.003	0.9544
BC	0.04	1	0.04	0.0008	0.9775
BD	28.19	1	28.2	0.55	0.4685
CD	18.57	1	18.58	0.36	0.5550
A^2	0.06	1	0.056	0.001	0.9741
B^2	194.96	1	194.96	3.84	0.0703
C^2	221.22	1	221.22	4.36	0.0556
D^2	64.15	1	64.15	1.26	0.2799
Residual	710.90	14	50.78		
Lack of Fit	657.25	10	65.72	4.9	0.0697
Pure Error	53.65	4	13.41		
Cor Total	1465.61	28			

Normal plots:

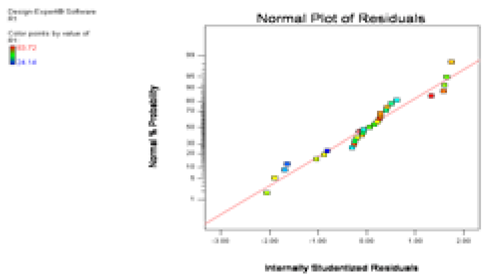

Figure.1.Normal plot for activated carbon

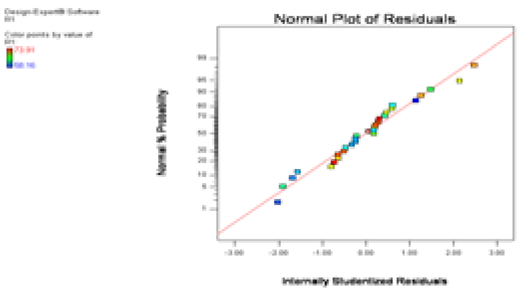

Figure.2.Normal plot for Green algae

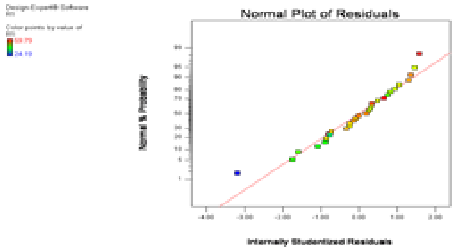

Figure.3.Normal plot for Red and brown algae

Predicted plots:

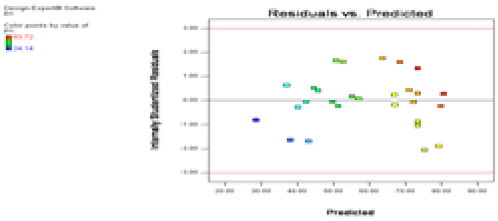

Figure.4.Predicted plot for activated carbon

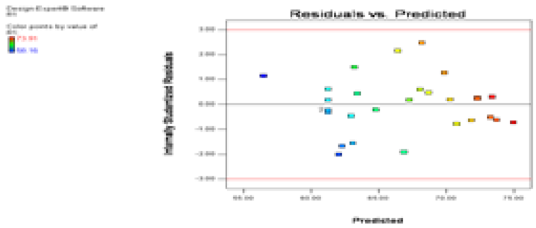

Figure.5.Predicted plot for Green algae

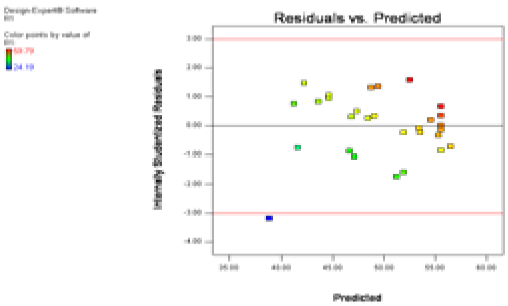

Figure.6.Predicted plot for Red and brown algae

CONCLUSION

Activated carbon, green algae and mixed red and brown algae were used as a adsorbent for the removal of xylene cyanol dye. The ranges and levels of three process variables were determined with the OFAT method. Response Surface methodology (RSM) was successfully applied to determine optimal conditions for maximum removal of Dye by using Box-Behnken design (Design Expert Software version 8.0). A quadratic regression model was proposed to describe the effects and interactions among influencing independent variables. The optimal conditions for maximal dye uptake for activated carbon; 200µg/mL of dye concentration,1.5g of dose, pH5,temperature 50ºC and incubation time of four hours yielding an experimental response of 82.40 and for green algae, 200µg/mL of dye concentration,0.5g of dose, pH4,temperature 70ºC and incubation time of six hours yielding an experimental response of 64.8,and for mixed red and brown algae200µg/mL of dye concentration,0.5g of dose, pH8,temperature 30ºC and incubation time of two hours yielding an experimental response of 45.90. The experimental model was found to be in agreement with the experimental value for all three adsorbates.

We tried for the first time to compare the dye uptake ability of three different adsorbates and optimized the conditions using RSM based statistical designs, but further validation using analytical approach is required to support/to predict the insights of mechanism sorption using commercially available and largely produced carbon.

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