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Removal of sunset YellowAzo dye using activated carbon entrapped in alginate from aqueous solutions

Abstract

The aim of this research is for decreasing contaminants using alginate beads with activated carbon entrapped (AG-AC) by adsorption. Different sunset yellow FCF concentrations were prepared in the laboratory. The effect of the different parameters was studied (pH, contact time, adsorbent dose, stirring rate, and concentrations) on the removal processes. The results were analyzed according to the Langmuir, Temkin, Freundlich, and Dubinin-Radushkevich adsorption isotherms. The removal efficiency is more appropriate by the Langmuir isotherm. Sunset yellow FCF (10 ppm) percent removal efficiency is (86%) at pH 3 and 10 g\L of the adsorbent dose for 30 min with a fixed stirring rate (100 rpm). The effect of different operating parameters was investigated using Linear regression analysis which occupies more than 97 % of the total of the variables affecting the removal process. Finally, I recommend using this technology for Removal of Sunset Yellow FCF.

Keywords: entrapped AG-AC, removal of sunset yellow FCF, statistical and isotherm studies

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Introduction

Sunset yellow FCF, is an azo dye entitled to as a food additive, which is used as a colorant for tablets, capsules, and cosmetics pharmaceuticals.[1–3] In most of the purpose, more than 50% of the dye is lost in wastewater.[4–6] Dyes are discharged into the ecosystem, causes to deterioration of water quality and by influencing plants and animals, and causes chronic and severe toxicity, and are a potential hazard to human.[5,7] For this reason, safety data, such as acceptable daily intake, based on toxicological studies on experimental animals and human clinical studies have been determined and evaluated by the Food and Agricultural Organization and World Health Organization.[8–10]

In view of these concerns, there have been many researchers across the earth evaluating the fate and removal of these contaminants in wastewater treatment processes as the classical treatment procedures are not efficient in removing the chemicals to levels below their potentially no effect concentrations.[11,12] Because of economic and technical drawbacks, sunset yellow FCF recycling and removal technologies have not yet been widely adopted.[13–15] Therefore, researchers have been focused on the modification process that more convenient in pollutants removal from wastewater.[16–19]

Activated carbon has a high adsorption capacity, but its cost in wastewater treatment is high because of the dispersion of the powder.[12,20] To overcome this problem and increasing the efficiency of the removal through entrapment activated carbon with alginate which helps in removal process where allows the contaminated aqueous medium to pass through it and be in contact with activated carbon.[19–21] It has become one of the best economics and effective wastewater treatment technique, this method has agitated great concern during the latest years.[20,22] The aim of this research is to explore the possibility of alginate beads with entrapped activated carbon for the removal of sunset yellow FCF from aqueous medium. In supplement, the Langmuir, Temkin, Freundlich, and Dubinin-Radushkevich adsorption isotherms are used to fit the data.

Experimental

Chemicals and reagents

All chemicals used were of the analytical reagent grade and of highest purity; such as activated carbon, sunset yellow FCF [$C_{20}H_{24}O_2$, Zhejiang Xianju], sodium alginate [($C_6H_7NaO_6$) n, Qingdao Bright Moon], sodium hydroxide [NaOH, Riedel-de Haën], hydrochloric acid [HCl, Scharlau], sulfuric acid [H_2SO_4, sever biotech], ethanol 96 % [C_2H_5OH, World co. for sub & med industries] and calcium chloride [$CaCL_2$, Fisher Scientific]. The change of pH was adjusted using 0.1 M NaOH and 0.1 M HCl solutions.

Methods

Preparation of Adsorbent: Alginate beads with entrapped activated carbon were prepared by adding one g of activated carbon into 2% (wt./v) of sodium alginate solution. The mixture solution was added drop by drop to a 5 % (wt./v) $CaCl_2$ solution. The adsorbent formed taken out and washed with distilled water to be used in the adsorption method.

Batch adsorption studies: Alginate beads with entrapped activated carbon were added to an aqueous sunset yellow FCF solution (5 ppm). Using different operating parameter: Effect of factors such as (pH, dose, concentrations, stirring rate, and contact time) was mixed with aqueous sunset yellow FCF solution, filter solution through fiber filter paper (WHATMAN 1441-125) and take specific amount of filtrate and reagents carefully in volumetric flask, then sunset yellow FCF evaluated according to 25th Edition of Standard Methods for the Examination of Water and Wastewater. The removal % was calculated using the following equation:

$$Sorption[\%]=[Co-Ce/Co]\times 100$$

Where C_o is the initial concentration (ppm) of sunset yellow FCF in solution and C_e is the equilibrium concentration (ppm) of sunset yellow FCF in solution. The amount of sunset yellow FCF adsorbed by Alginate beads with entrapped activated carbon was calculated using the following equation:

$$qe\left[mg/g\right] = \left[\left[Co - Ce\right]V\right]/\ m$$

where V is the volume of aqueous medium (L), q_e is the equilibrium adsorption capacity (mg/g), and m is the dry weight of the adsorbent (g).

Adsorption study

Freundlich isotherm: The Freundlich isotherm model[20,23] is an empirical equation employed to describe heterogeneous adsorption surface and is given by:

$$Ln\,qe = 1/n\,ln\,Ce + ln\,Kf$$

Where $K_f\left((mg/g)\ (mg/L)^{-1/n}\right)$, and n (dimensionless) are Freundlich constant related to the adsorption intensity and adsorption capacity, respectively. (K_f) and (n) evaluated by plotting ln q_e and ln C_e.

Langmuir isotherm: Langmuir isotherm model supposes a monolayer coverage of adsorbate over a homogeneous adsorbent surface.[12,24] The Langmuir linearized is given by the equation:

$$Ce / qe = 1 / (KL\ qmax) + Ce/q\ max$$

Where C_e (mg/L) is equilibrium concentration of sunset yellow FCF, q_e (mg/g) is the mass of sunset yellow FCF adsorbed per mass of adsorbent used, K_L (L/mg) is the Langmuir constant related to binding sites affinity and adsorption energy, and q_{max} (mg/g) is the maximum monolayer capacity of adsorption. The plot of C_e/q_e versus C_e employed to generate the values of q_{max} and K_L.

Temkin isotherm: The Temkin isotherm study has been chosen to estimate the adsorption potential of the adsorbed and adsorbent solution. This model contains a factor which takes into respect the interactions of the membrane (adsorbent-adsorbate) and the aqueous solution ions.[19,25] The Temkin isotherm model has been generally applied in the following equations:

$$Qe = (RT/b)\ Ln\ (AT\ Ce)$$

$$Qe = (RT/bT)\ Ln\ AT + (RT/b)\ Ln\ Ce$$

$$B = RT/bT$$

A linear expression of the Temkin equation is represented by:

$$Qe = B\ Ln\ AT + (RT/b)\ Ln\ Ce$$

where:

A_T = the equilibrium binding constant [L/g], b_T = the adsorption constant [J/mol K], R = universal gas constant (8.314 J/mol K), T = absolute temperature value [298 K], B = a constant related to the heat of sorption [J/mol].

Dubinin-Radushkevich isotherm: Dubinin–Radushkevich isotherm model is generally applied to expresses the adsorption mechanism with the distribution of Gaussian energy onto a heterogeneous surface.[19,26] The isotherm model has often successfully fitted the intermediate range of concentrations data well and high solute activities.

$$Qe = B\ Ln\ AT + (RT/b)\ Ln\ Ce$$

$$Qe = B\ Ln\ AT + (RT/b)\ Ln\ Ce\ Ln\ Qe = Ln\left(Qs\right)\_\ (K_{ad}\ \varepsilon^2\ )$$

Where Q_e, Q_s, K_{ad}, are Q_e = adsorbate amount in the adsorbent at equilibrium (mg/g); Qs = capacity of theoretical isotherm saturation (mg/g); K_{ad} = Dubinin–Radushkevich isotherm constant (mol^2/kJ^2) and Ɛ=Dubinin–Radushkevich isotherm constant. The approach was applied to distinguish the chemical and physical adsorption of metal ions with its mean free energy, E per molecule of adsorbate can be computed by the relationship.

$$E=[1/(\sqrt{}\ 2BDR)]$$

Where B_{DR} is denoted as the isotherm constant.

Results and discussion

Effect of pH

The influence of pH solutions value on the amount of sunset yellow FCF removed by Alginate beads with entrapped activated carbon from the aqueous medium was evaluated by carrying out experiments with different pH values (3, 5, 7 and 9) at different contact times (30, 45 and 60 min), with removal efficiencies of (86, 68, 42 and 26%), (87, 69, 45 and 27 %) and (88, 70, 46 and 28 %) respectively, and plots of the pH solution against the removal percentage of the sunset yellow FCF that was removed from the solution were shown in Figure 1. The conditions used were: the sunset yellow FCF concentration was 10 ppm, the adsorbent dose 10 g\L, and the stirring rate was fixed at 100 rpm. The optimum pH value for the removal was 3. The pH solution plays an important role in affecting sunset yellow FCF adsorption. In acidic pH, the surface of adsorbents become protonated so the amount of dye adsorbed increase due to the electrostatic attraction between the positively charged surface and the negatively charged sunset yellow FCF molecules. On the hand, in the alkaline pH, a negatively charged surface sites on the adsorbent didn't favor the adsorption of sunset yellow FCF anions due to electrostatic repulsion. Similar results were reported in the scientific manuscripts for the adsorption of sunset yellow FCF.[27]

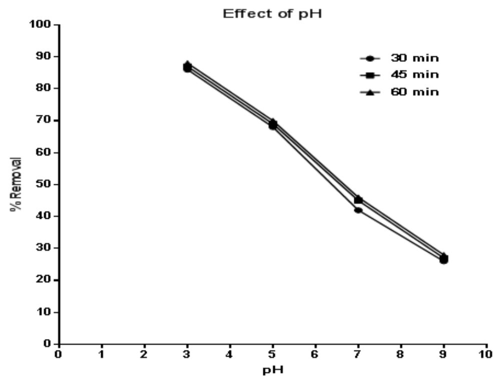

Figure 1 Effect of pH on Sunset yellow FCF removal.

Effect of contact time

Figure 2 depicts the removal efficiency as a function of contact time. The effect on sunset yellow FCF removal studied at different times (15, 30, 45, 60, 120 and 180 min) using 10 g\L of the adsorbent dose at pH 3, and the stirring rate was fixed at 100 rpm. Sunset yellow FCF concentration was (10 ppm) and the removal percentages were (81, 86, 87, 88, 90 and 90 %). From the data given from Figure 2, it is shown that, increased removal efficiency with increase in contact time. As shown, sunset yellow FCF uptake by entrapment activated carbon with alginate was very rapid at the first 15 min. After the first

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15 min, the sunset yellow FCF uptake progressively decreased with time. As the treatment time proceeded, the adsorbent available sites had the inclination toward saturation. Equilibrium was established after 30 min. Increasing in time leads to an increase in the contact between the contaminant to the larger surface area of adsorbent as there are many adsorption sites. Similar results were reported in the scientific manuscript for the adsorption of sunset yellow FCF.[3,28]

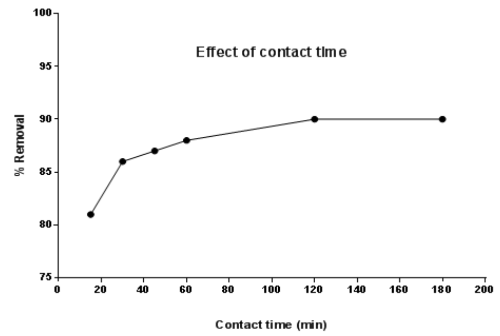

Figure 2 Effect of contact time on Sunset yellow FCF removal.

Effect of adsorbent dose

Figure 3a depicts sunset yellow FCF removal efficiency is related to adsorbents dose. The adsorbent doses were varied between 5 and 25 g/L, other operational factors such as pH, contact time, concentration and stirring rate were 3, 30 min, 10 ppm, and 100 rpm, respectively. The optimum dose for sunset yellow FCF removal was about 10 g\L, and the removal percentages were (83, 86, 88, 92, 95 %), as shown in Figure 3b, respectively. As expected, at high adsorbent does the removal increased because of the increased adsorbent surface area and the number of available vacant adsorption sites increased. Similar results were reported in the scientific literature for the adsorption of sunset yellow FCF.[29,30]

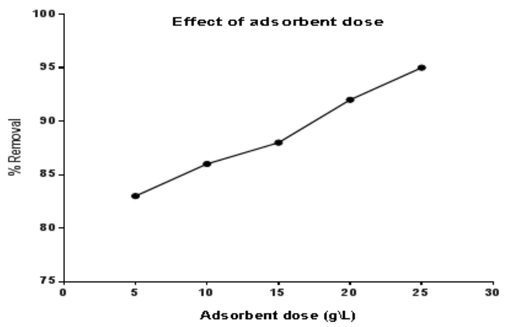

Figure 3a Effect of adsorbent dose on Sunset yellow FCF removal.

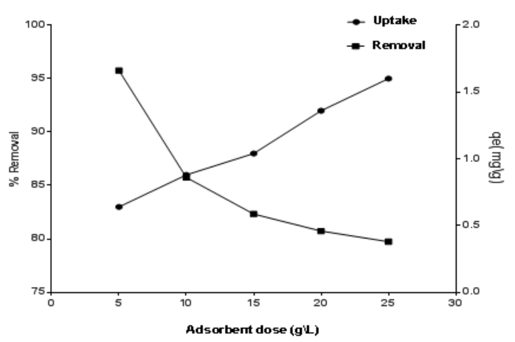

Figure 3b The optimum effective dose for Sunset yellow FCF removal.

Effect of stirring rate

Figure 4 depicts sunset yellow FCF removal efficiency by the adsorbent as a function of stirring rate. The stirring rate was varied between 100 and 500 rpm. The other operational factors such as pH, contact time, concentration, and adsorbents dose were 3, 30 min, 10 mg/L, and 10 g\L, and the removal percentages were (86, 87, 88, 89 and 89 %), as shown in Figure 4. The optimum stirring rate for removal was 100 rpm. The increasing in stirring speed resulting to increase in sunset yellow FCF removal efficiency, due to the fact that, increasing in stirring rate enhanced the contaminants diffusion of sunset yellow FCF on the surface of the adsorbent. Similar results were reported in the scientific manuscript for the adsorption of sunset yellow FCF.[31]

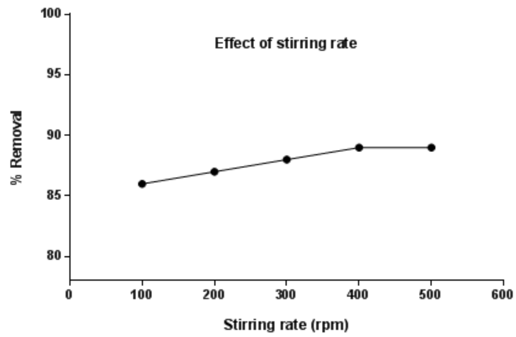

Figure 4 Effect of stirring rate on Sunset yellow FCF removal.

Effect of the concentration

The effect of concentration of the aqueous solution on the percent sunset yellow FCF reduction by the adsorbent was studied at various concentrations varying from (10 to 50 ppm) were shown in Figure 5a. Other operational factors (pH, contact time, stirring rate and adsorbents dose) were 3, 30 min, 10 rpm, and 10 g\L, and the removal percentages were (86, 83, 79, 76 and 71 %), as shown in Figure 5. It can be observed that adsorption was lower at higher concentrations of sunset yellow FCF and vice versa. Similar results were reported in the scientific literature for the adsorption of sunset yellow FCF.[2,32]

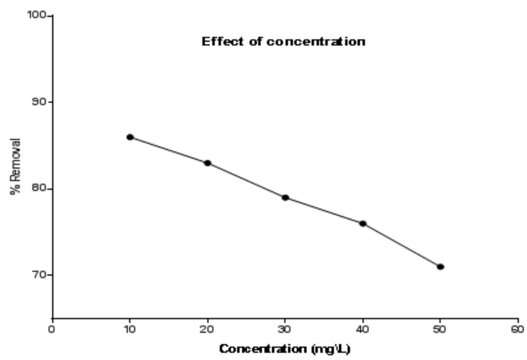

Figure 5a Effect of concentration on Sunset yellow FCF removal.

Adsorption isotherm study for Sunset yellow FCF removal

The sorption capacity of the adsorbent predicted and evaluated by adsorption study. The adsorption model equilibrium data obtained at different sunset yellow FCF concentrations were described using Langmuir, Freundlich, Dubinin-Radushkevich (D–R), and Temkin equations. They are the most four common adsorption isotherms applications used for wastewater treatment technique, under various conditions of pH, concentrations, adsorbent dose, contact time and stirring rate.[33] The suitability and acceptability of the adsorption isotherm equations to the equilibrium data were based on the values of the correlation coefficients, R^2 estimated from linear regression of the least square fit statistic on Micro Math Scientist software**.** Figure 5(b-e) represent the adsorption isotherms model of sunset yellow FCF. The adsorption data were fitted well with the Langmuir isotherm with the highest (R^2 = 0.997), as shown in Table 1. The n value > 1 from Freundlich, and maximum adsorption capacity of 5.40 mg/g from Langmuir with K_L < 1 obtained are the indication that the adsorption is favorable on the investigated adsorbent (Table 1).

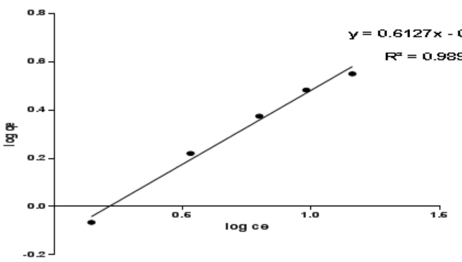

Figure 5b Freundlich adsorption isotherm for Sunset yellow FCF contributing component.

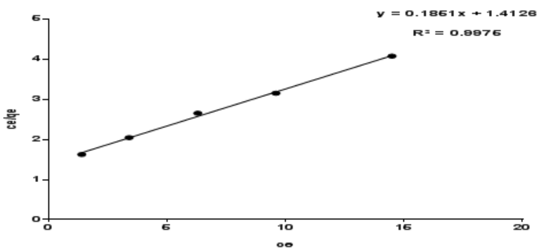

Figure 5c Langmuir adsorption isotherm for Sunset yellow FCF contributing component.

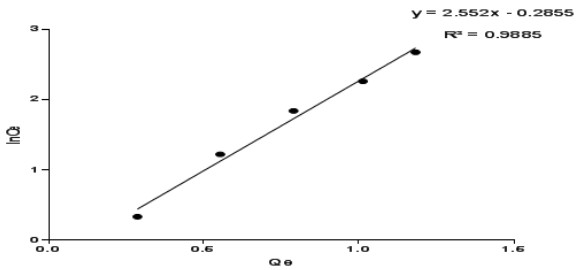

Figure 5d Temkin adsorption isotherm for Sunset yellow FCF contributing component.

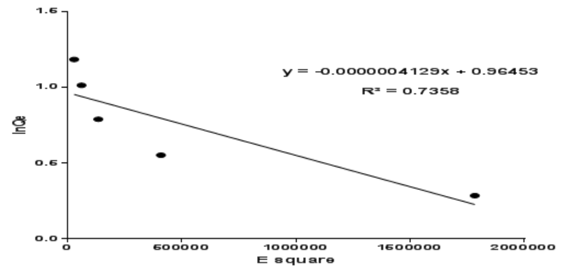

Figure 5e Dubinin–Radushkevich adsorption isotherm for Sunset yellow FCF contributing component.

Table 1 Isotherm parameters for the adsorption of Sunset yellow FCF

Isotherm name	Parameters	Values
Freundlich	K_f (mg/g(mg/L)$^{1/n}$)	1.35
	n	1.63
	R^2	0.9894
Langmuir	Qmax (mg/g)	5.40
	K_L (L/mg)	0.131
	R^2	0.9976
Temkin	A_T (L/g)	1.12
	b_T (J/mol g/mg)	970.99
	B	2.55
	R^2	0.9886
Dubinin–Radushkevich	Qs (mg/g)	2.62
	K_{ad}	4.1E-07
	R^2	0.7368

Statistical analysis

The effect of the following variables on the removal process e.g. pH, contact time, dose, concentration, and stirring rate have been studied using the entered method, where it was found that R^2 = 0.971, as shown in Table 2b. This means that the studied variables profane occupy more than 97 % of the total of the variables affecting the removal process as that the standard error of the estimate is 2.81477, which means that the percentage of error in this study is very low. ANOVA program was applied and the data were given in Table 2c. This table showed the sum of squares and the effect of the whole model and it was observed that the P value (0.000) where the model is considered successful if P value less than 0.05.

Table 2a Variables Entered/Removed[a]

Model	Variables entered	Variables removed	Method
1	concentration, dose, time, pH, stirring b		Enter

a. Dependent Variable: removal
b. All requested variables entered

Table 2c ANOVA [a]

Model	Sum of squares	df	Mean square	F	Sig.
Regression	5114.505	5	1022.901	129.107	0.000b
Residual	150.535	19	7.923		
Total	5265.04	24			

a. Dependent Variable: removal
b. Predictors: (Constant), concentration, dose, time, pH, stirring

Table (2d) [Coefficients[a]]: The data given in Table 2d showed the sum of squares, P value, and T value for each variable. From the table, it can be inferred that all variables had an effect on the removal process except (contact time, adsorbent dose and stirring rate), where the P value is larger than 0.05 where this means that it can be neglected during the removal process.

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Table 2d Coefficients[a]

Model	Unstandardized coefficients		Standardized coefficients	t	Sig.
	B	Std. error	Beta		
(Constant)	118.763	3.139		37.838	0
pH	-9.958	0.444	-0.973	-22.434	0
time	0.024	0.019	0.056	1.261	0.223
dose	0.037	0.081	0.022	0.455	0.654
stirring	0.006	0.006	0.043	1.005	0.327
concentration	-0.382	0.061	-0.268	-6.262	0

a. Dependent Variable: removal

The effect of different operating parameters was investigated using Linear regression analysis using statistical algorisms where the obtained results support the practical results. By applying the B values shown in the table, the removal equation can be deduced whereas:

Where X_1 is the effect of pH (3, 5, 7 and 9), X_2 is the effect of contact time (15, 30, 45, 60, 120 and 180 min), X_3 is the effect of adsorbent dose (5, 10, 15 20 and 25 g/L), X_4 is the effect of stirring rate (100, 200, 300, 400 and 500 rpm) and X_5 is the effect of concentration (10, 20, 30, 40 and 50 mg/L).[34,35]

Conclusion

In this study, the sunset yellow FCF concentration can be eliminated by entrapment activated carbon with alginate biopolymer. Various operating factors on the efficiency of sunset yellow FCF removal investigated and optimized. Removal affected by the experimental parameters such as contact time, dose, pH, stirring rate, concentration. Maximum sunset yellow FCF removal was observed at the pH 3. Sunset yellow FCF removal efficiency between (86 and 71 %) was achieved after using different sunset yellow FCF concentrations (10, 20, 30, 40 and 50 ppm) with 10 g\L of the adsorbent, stirring rate 100 rpm, and contact time 30 min. When the entrapped activated carbon in alginate biopolymer dose was increased from 5 to 25 mg/L, the removal of sunset yellow FCF increased by 12 % (C_0 = 10 mg/L). The adsorption data were fitted well with the Langmuir isotherm with the highest (R^2 = 0.997). The entrapped activated carbon in alginate biopolymer is a cost-effective alternative and can lead to success in wastewater treatment and produce high-quality treated effluent.

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None.

Conflicts of interest statement

The authors whose names are listed certify that they have NO affiliations with or involvement in any organization or entity with any financial interest (such as honoraria; educational grants; participation in speakers' bureaus; membership, employment, consultancies, stock ownership, or other equity interest; and expert testimony or patent-licensing arrangements), or non-financial interest (such as personal or professional relationships, affiliations, knowledge or beliefs) in the subject matter or materials discussed in this manuscript.

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