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ADSORPTION OF YELLOW 5 GL DYE USING ACTIVATED CARBON PREPARED FROM THE SEEDS OF SUGAR APPLE (*ANNONA SQUAMOSA* L.)

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ABSTRACT: The study was carried out for the removal of Yellow 5 GL dye using seeds of Sugar Apple (*Annona squamosa* L.). The activated carbon prepared from seeds of Sugar Apple was obtained from the local market. The activated carbon prepared was subjected to thermodynamics, kinetics and isotherm studies. Batch adsorption studies were conducted to assess the adsorption efficiency of the adsorbent over the adsorbate. The negative value of change of standard free energy (ΔG^0) confirming the thermo dynamic feasibility and spontaneous nature of the process and positive values of change of standard enthalpy and standard entropy showed the process was endothermic and increased randomness at the solid-liquid interface. The equilibrium data fitted to Langmuir isotherm revealed monolayer adsorption and kinetic data fitted to pseudo-second order.

Keywords: Dye, Isotherm, Spontaneous, Adsorption.

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1. INTRODUCTION

Pollution of water due to municipal, industrial, mining, agricultural and other human activities is one of the most serious problems faced by developed as well as developing countries in recent years. The textile industry is considered to be one of the biggest threats to the environment. This is mainly due to the fact that colored effluents are composed of non-biologically oxidisable component because of the molecular size and structure of the dye-stuff [1]. The water let out after the production

of textiles contains a large amount of dyes and other chemicals. Several techniques have been adopted for water and wastewater treatment[2,3] such as membrane filtration [2], coagulation/flocculation[3,4], adsorption[5], ion-exchange[6-10], oxidation[11-13] , flotation[2,3], chemical reduction[14], biological treatment etc. However, these developed methods are varying in their effectiveness, cost and environmental impact. Therefore, a suitable cheapest removal technology is needed. Adsorption technique is very simple in design [15], initial cost, and ease of operation and insensitive to toxic surface. A large number of adsorbents such as activated carbon [16], polymeric resin, modified and unmodified low cost adsorbents to remove dye. As such there has been increasing in research into cheaper and more readily available adsorbents. Adsorption by activated charcoal has been found to be promising techniques for dye wastewater treatment because of its low cost, simplicity, high efficiency and wide-ranging availability for removing dyes. However, there are certain drawbacks associated with its use especially the expensive nature of high quality activated carbon. These adsorbents were found to be efficient in binding with basic dyes rather than acid dyes. However some of these adsorbents do not have good adsorption capacities for the anionic dyes because most have hydrophobic and anionic surfaces. Hence there is a need to search for more effective adsorbents to remove acid dyes. The present study was investigated for the adsorption of Yellow 5 GL an acid dye on the activated carbon prepared from the seeds of Sugar Apple (ASC). The effect of pH, adsorbent dose, temperature and initial dye concentrations were studied for the removal of dye. Discharge of dye bearing waste water makes an adverse effect on aquatic environment because the dyes give water undesirable color[17] and reduce light penetration and photosynthesis [18-20]. In comparison with other techniques adsorption is superior in simplicity of design, initial cost, ease of operation and insensitivity to toxic substances. This technique uses a large number of suitable sorbents as activated carbon [21, 22], polymeric resins [22-25], or various low cost adsorbents (non-modified or modified cellulose biomass, chitin, soil material, activated alumina, bacterial biomass etc.,) [26-30].

2. MATERIALS AND METHODS

The seed of sugar apple was obtained from the local market. It was cleaned dried and ground to powder. It was soaked in 1:1 sulphuric acid for 48 hrs and activated at 160^0 C for 6 hrs. The activated carbon thus obtained was washed with distilled water till the pH of the washed water became neutral and then dried. The dried activated carbon was passed through different sieve plates for getting particle sizes in the range of 75 to 150 μm. The activated carbon prepared was stored for later use.

Preparation of dye solution

0.5 g of Yellow 5 GL dye was weighed and dissolved in 1L of double distilled water to get a standard solution of 500 mg/L. Working solutions of desired concentrations were prepared by succeeding dilution.

Batch adsorption experiments

Batch experiments were carried out to study the adsorption efficiency of the adsorbent. The studies were performed in a 100 ml iodine flask by mixing 50 ml of different initial concentrations (100 mg/L, 150 mg/L, 200 mg/L) with 50 mg of the adsorbent. The mixture was agitated at 130 rpm in rotary shaker for pre-determined time. After equilibrium was attained the mixture was filtered and the adsorption of the dye solutions was measured using UV/Visible spectra photometer. The amount of adsorbate adsorbed at equilibrium was calculated using the equation.

$$qe = \frac{Co - Ce}{M} xV$$

q_e is the amount of dye adsorbed at equilibrium (mg/g) where Co and Ce are the liquid phase concentrations of adsorbate) at initial and equilibrium concentration (mg/L) respectively. M is the weight of the adsorbent used (g) and V is the volume of solution (ml)

The batch experiments for the adsorption kinetics were similar to that of equilibrium studies.

Adsorption thermodynamics and equilibrium isotherms

The thermodynamic parameters such as change in standard Gibbs free energy (ΔG^0) were calculated from Vant Hoff's expression. The equilibrium data were analyzed using Langmuir, Freundlich and Temkin isotherms. The possibility of intra particle diffusion was investigated using the expression described big Weber and Morris (1963) as presented in Equation.

$$Qt = kidt ½ Ci$$

The isotherm provides relationship behaviors of concentration of the dye in solution and the amount of dye adsorbed on the solid phase when both phases are in equilibrium. The equilibrium of experimental data for adsorbed dye on adsorbent is studied by using the Langmuir, Freundlich and Temkin isotherm models. In this study the best fit isotherm model from the experimental data were determined using the value of correlation co-efficient (R^2).

3. RESULTS AND DISCUSSION

Influence of pH

pH is one of the most important parameters influencing the adsorption process. The pH of the solution primarily affects the degree of ionization of the dye and the surface properties of the adsorbents. The figure 1 shows the effect of p^H on the percentage removal of dye. The maximum adsorption was observed at pH 3. Yellow 5 GL dye is an anionic dye, the positive charge density at low pH attracts the dye. A slight decrease at pH 2 might be due to the protonation of the sulphonate and aliphatic amino group of the dye.

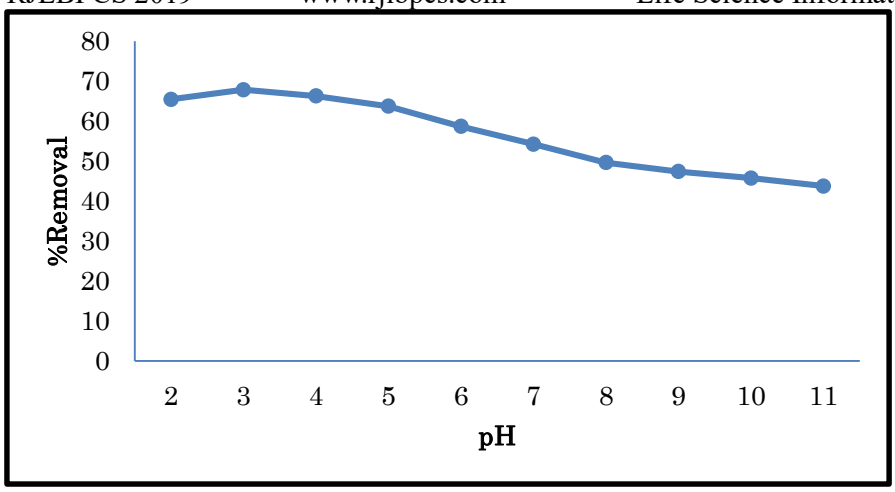

Figure 1: Effect of pH on the % of Removal of Yellow 5GL dye

Influence of Temperature

Figure 2 represent the influence of temperature on the percentage removal of dye at a given concentration. The temperature has great effect on the adsorption process. The percentage of adsorption increase with increased of temperature might be due to the increase of intra particular diffusion rate of adsorbate ions into the pores at a higher temperature suggesting an endothermic process.

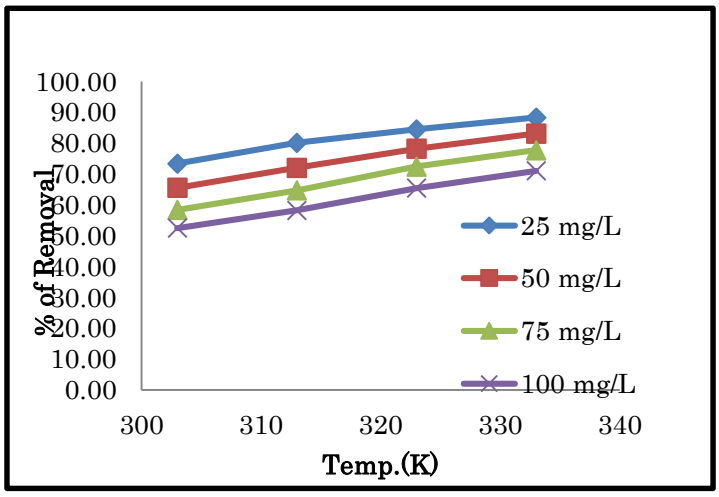

Figure 2: Temperature on the % of Removal for Yellow 5 GL dye

Influence of contact time

The percentage removal of dye as a function of contact time was studied. It was understood from the figure 3 that the percentage removal of dye increased steadily at the beginning and after sometimes it reached the equilibrium at all initial concentrations. This could be explained on the basis that at the beginning plenty of vacant active surface site available for the adsorption but as the time increased the number of active vacant surfaces decreased and at the same time the repulsive forces between the absorbate and the adsorbent increased.

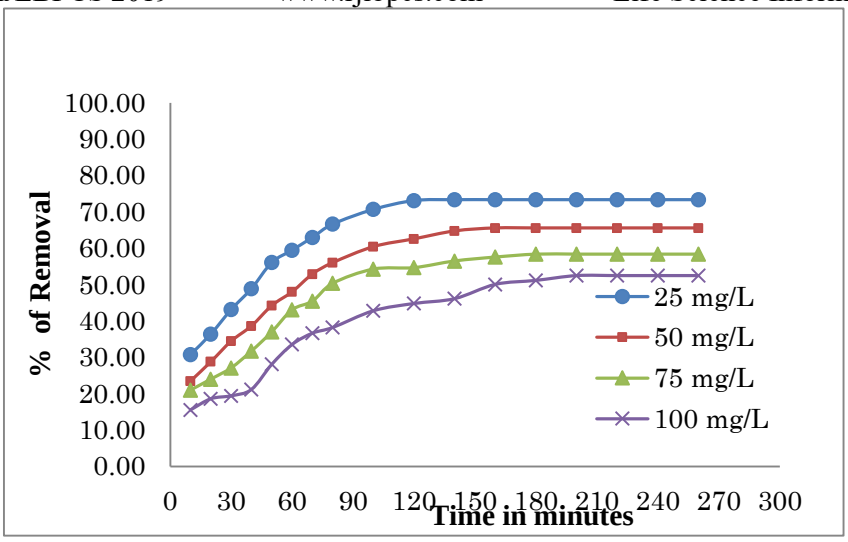

Figure 3: Effect of contact time on the Ci for Removal of Yellow 5 GL dye

Influence of adsorbent dose

It was observed from the figure (4) that the percentage removal of dye increased with increase of adsorbent dose at pH 3 and temperature 303 K and at a given initial concentration. Since the surface area increases with increase of adsorbent dose naturally the adsorption of the dye on the surface of the adsorbent increases.

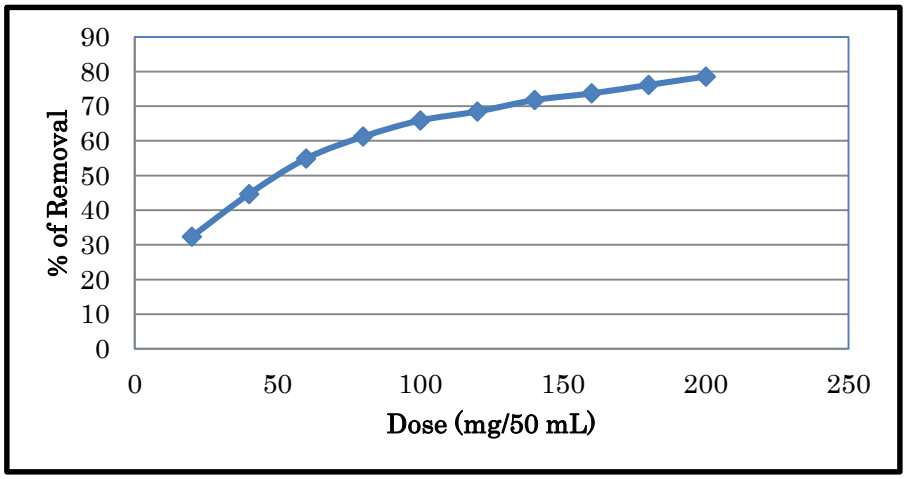

Figure 4: Effect of Dose vs % of Removal for Yellow 5 GL dye ions

Influence of initial concentration

The influence of initial concentration of the dye on the percentage removal at different temperatures was debited in the figure (5). It was observed that the percentage removal decreased with the increase of initial concentration for a given amount of an adsorbent. The percentage removal of the dye increased with the lower initial concentration might be due to the availability of sufficient numbers of adsorption sites initially. But at higher concentration the number of available adsorption sites was low and hence the percentage removal became depend on initial concentration. At a given initial concentration the percentage removal increased with an increase of temperature suggested the adsorption is endothermic.

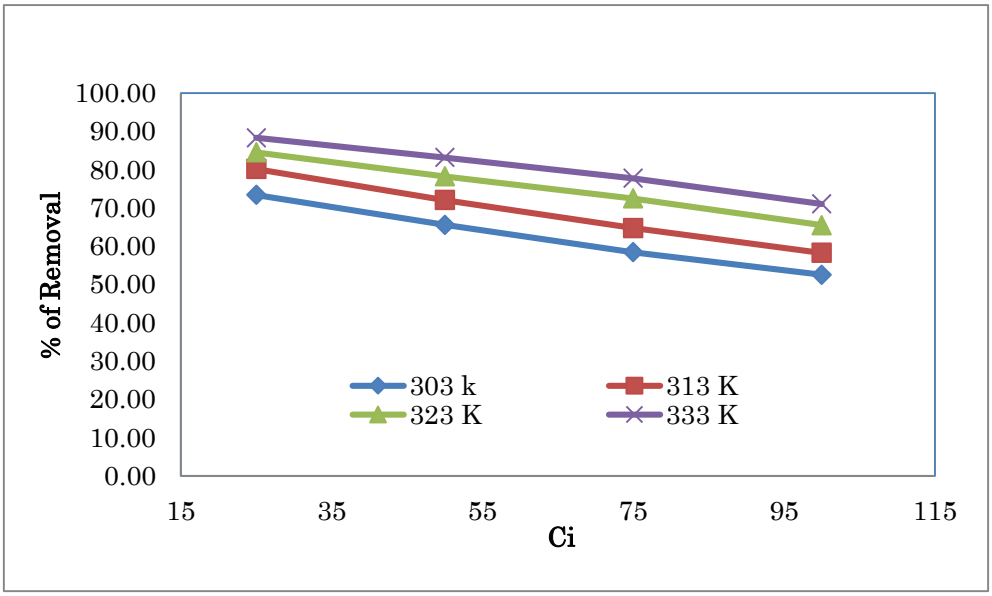

Figure 5: Effect of Ci vs % of Removal for Yellow 5 GL dye ions

Thermodynamic study

The results of thermodynamic parameters calculated for the adsorption of the dye on the adsorbent shown in the table and figure. The ΔG^0 showed negative that implied adsorption was thermodynamically feasible and spontaneous in nature. The possible values of ΔG^0 and ΔS^0 favoured the reaction was increased randomness at the solid liquid interface.

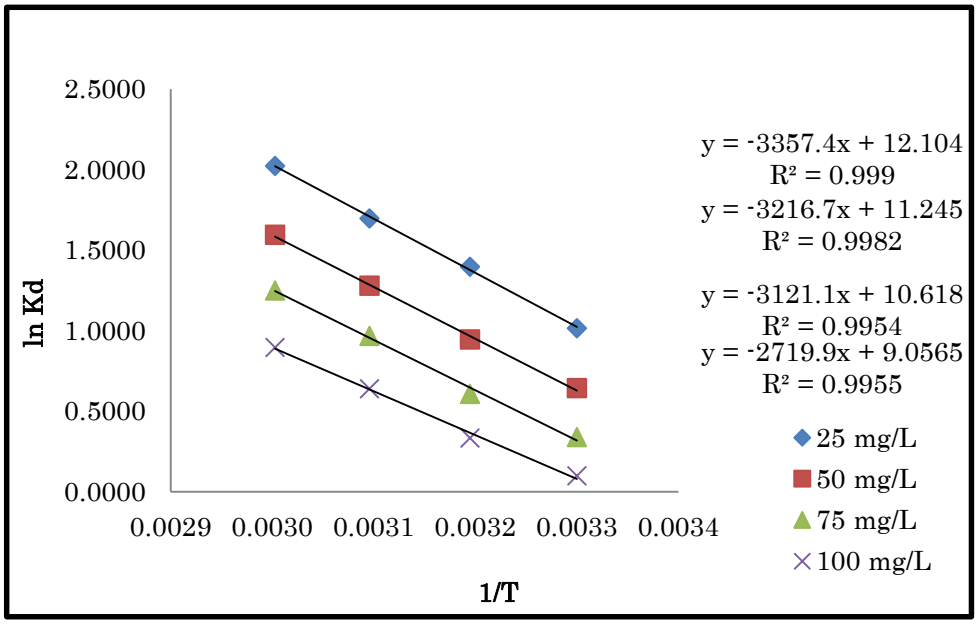

Figure 6: Van't Hoff Plot for Yellow 5 GL dye ions

Adsorption kinetics

The figures 7 and 8 showed the pseudo – first order (Lagergren's) and pseudo – second order (Ho's) plots of adsorption dye and are tabled in the table. The correlation coefficient (R^2) implied the pseudo- second order kinetic model for the adsorption. Same results also have been reported in literature [31,32].

Sorption Isotherms

The experimental equilibrium sorption data were analyzed using three adsorption isotherm models Freundlich [33], Langmuir [34] and Temkin [35] isotherms.

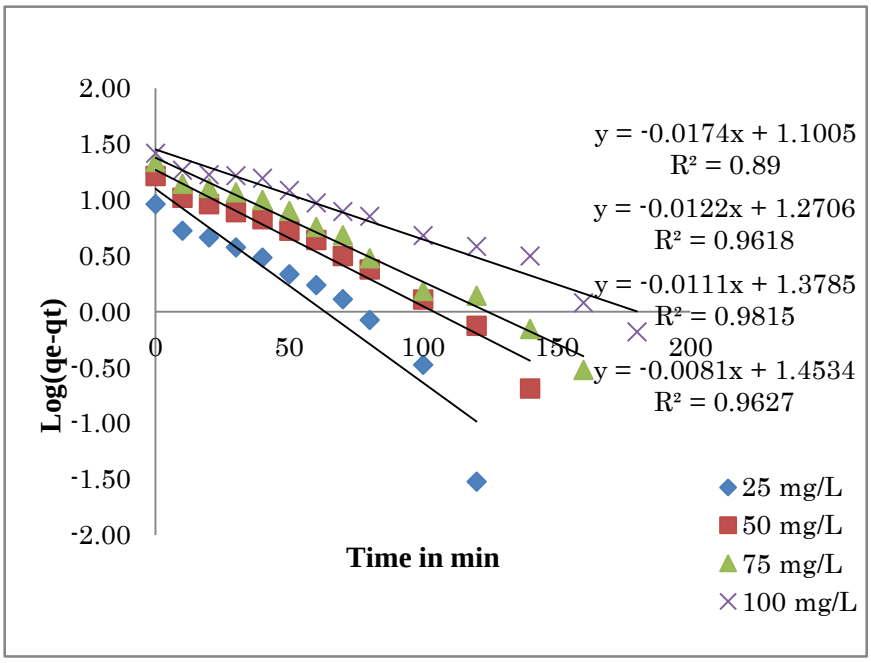

Figure 7: Lagergren plots for Yellow 5 GL dye ions

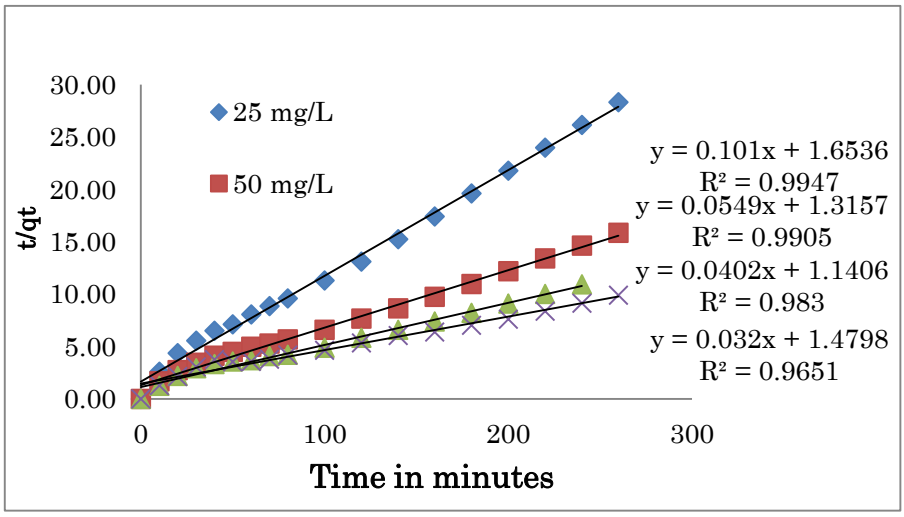

Figure 8: Ho plots for Yellow 5 GL dye ions

Equilibrium isotherm

The results of adsorption isotherm study was presented in the table 9,10,11 and showed in figure. The equilibrium data was best fitted Langmuir isotherm monolayer adsorption of the dye on the adsorbent.

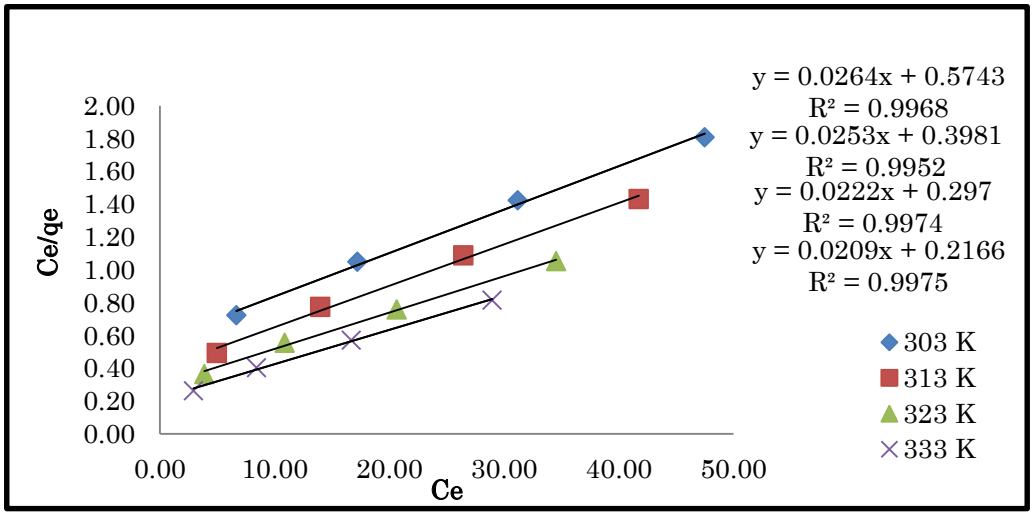

Figure 9: Langmuir isotherm constants for Yellow 5 GL dye ions

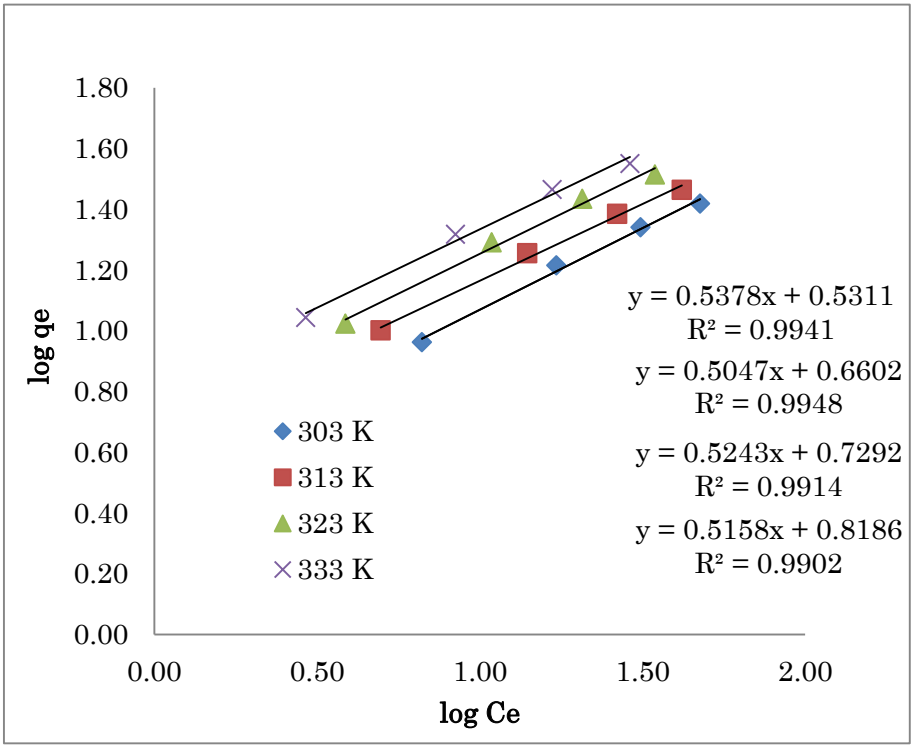

Figure 10: Freundlich isotherm constants for Yellow 5 GL dye ions

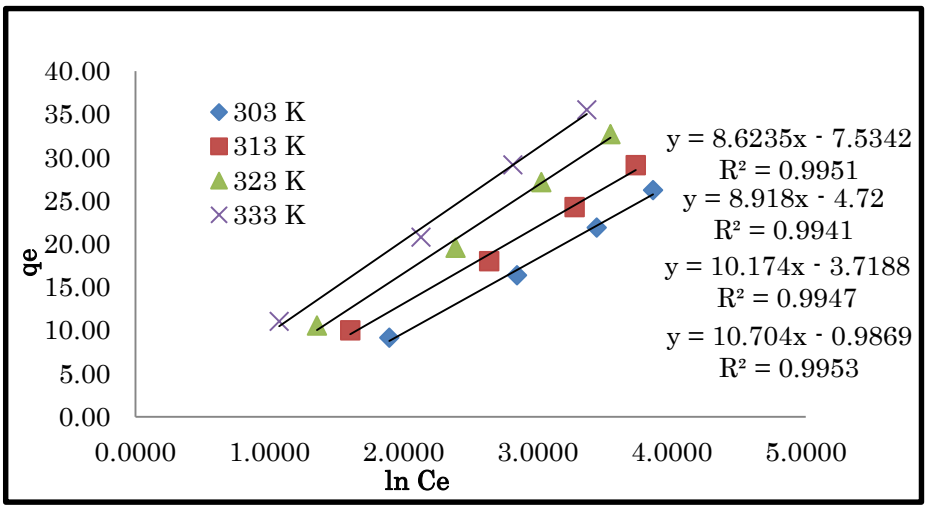

Figure 11: Temkin isotherm results for the adsorption of Yellow 5 GL dye ions

Table 1: Effect of pH Vs % removal of yellow 5GL dye

(Ci: 50 mg/L, Dose: 100 mg/50 mL, Time: 4 hrs, Temp: 303K)

pH	2	3	4	5	6	7	8	9	10	11
%R	65.46	67.87	66.32	63.76	58.72	54.25	49.67	47.43	45.78	43.76

Table 2: Effect of temperature Vs % removal of yellow 5 GL dye

(Dose: 100 mg/50 mL, Time: 4 hrs)

Ci	25				50				75				100			
Temperature	303	313	323	333	303	313	323	333	303	313	323	333	303	313	323	333
%R	74.4	78.75	85.4	85.75	70.37	75.5	79.57	82.93	66.98	71.93	76.45	80.33	62.8	68.08	72.34	76.76
q_e	9.92	10.5	40.99	11.43	14.07	15.1	15.91	16.59	17.86	19.18	20.39	21.42	20.93	22.69	27.11	25.59

Table 3: Effect of contact time vs % removal of yellow 5 GL dye

(Dose: 100 mg /50 mL, pH: 3, Temp: 303K)

Time (min)	C_i :25		C_i : 50		C_i :75		C_i : 100	
	%R	q_t	%R	q_t	%R	q_t	%R	q_t
10	17.30	30.80	23.50	5.88	59.23	21.03	15.60	7.80
20	15.89	36.44	28.82	7.21	56.98	24.03	18.66	9.33
30	14.21	43.16	34.48	8.62	54.65	27.13	19.46	9.73
40	12.76	48.96	38.66	9.67	51.18	31.76	21.22	10.61
50	10.97	56.12	44.26	11.07	47.21	37.05	28.13	14.07

60	10.12	59.52	48.04	12.01	42.67	43.11	33.57	16.79
70	9.24	63.04	52.92	13.23	40.89	45.48	36.68	18.34
80	8.33	66.68	56.04	14.01	37.21	50.39	38.24	19.12
100	7.32	70.72	60.44	15.11	34.28	54.29	42.84	21.42
120	6.71	73.16	62.58	15.65	33.98	54.69	44.83	22.42
140	6.65	73.40	64.76	16.19	32.60	56.53	46.17	23.09
160	6.65	73.40	65.58	16.40	31.80	57.60	50.09	25.05
180	6.65	73.40	65.58	16.40	31.20	58.40	51.18	25.59
200	6.65	73.40	65.58	16.40	31.20	58.40	52.50	26.25
220	6.65	73.40	65.58	16.40	31.20	58.40	52.50	26.25
240	6.65	73.40	65.58	16.40	31.20	58.40	52.50	26.25
260	6.65	73.40	65.58	16.40	31.20	58.40	52.50	26.25

Table 4: Effect of dose vs % removal of yellow 5 GL dye

(Ci: 50 mg/L, pH: 3, Time: 4 hrs., Temp: 303K)

Dose	20	40	60	80	100	120	140	160	180	200
%R	32.38	44.65	54.87	61.2	65.88	68.45	71.75	73.67	76.11	78.54

Table 5: Effect of initial concentration at different temperatures vs

% removal of yellow 5 GL dye

Temperature (K)	Initial Concentration mg/L)	Concentration at equilibrium	Percentage of removal	Quantity adsorbed at equilibrium (mg/g)
303	25	6.65	73.40	9.2
	50	17.21	65.58	16.4
	75	31.20	58.40	21.9
	100	47.50	52.50	26.3
313	25	4.96	80.16	10.0
	50	13.98	72.04	18.0
	75	26.45	64.73	24.3
	100	41.75	58.25	29.1
323	25	3.87	84.52	10.6
	50	10.88	78.24	19.6
	75	20.65	72.47	27.2
	100	34.53	65.47	32.7
333	25	2.92	88.32	11.0

	50	8.42	83.16	20.8
	75	16.69	77.75	29.2
	100	28.96	71.04	35.5

Table 6: Thermodynamic parameters for yellow 5 GL dye

C_i (mg/L)	Temperature (K)	K_d	ΔG° (kJ/mol)	ΔH° (kJ/mol)	ΔS° (J/K mol)
20	303	0.4408	-2.5574	27.9134	100.6
	313	0.6064	-3.6343		
	323	0.7372	-4.5592		
	333	0.8786	-5.6021		
30	303	0.2800	-1.6242	26.7436	93.5
	313	0.4110	-2.4634		
	323	0.5558	-3.4372		
	333	0.6936	-4.4222		
40	303	0.1473	-0.8547	25.9488	88.3
	313	0.2638	-1.5807		
	323	0.4203	-2.5992		
	333	0.5433	-3.4640		
50	303	0.0435	-0.2522	22.6132	75.3
	313	0.1446	-0.8668		
	323	0.2778	-1.7183		
	333	0.3897	-2.4848		

Table 7: Lagergren kinetic equation parameters for the adsorption of yellow 5 GL dye ions

Dye	C_i (mg/L)	k_1 (10^{-2} min^{-1})	$q_{e(cal)}$ (mg/g)	$q_{e(exp)}$ (mg/g)	$\|\Delta q_e\|$ (mg/g)	MSSE
Yellow 5 GL	25	4.01	10.1	9.2	0.9	0.96
	50	2.81	18.6	16.4	2.3	
	75	2.56	23.9	21.9	2.0	
	100	1.87	28.4	26.3	2.2	

Table 8: Kinetic equation parameters for the adsorption of yellow 5 GL dye ions

Dye	C_i (mg/L)	k_2 (10^{-3}g/mg.min)	$q_{e(cal)}$ (mg/g)	$q_{e(exp)}$ (mg/g)	$\|\Delta q_e\|$	MSSE
Yellow 5 GL	25	6.17	9.9	9.2	0.7	1.53
	50	2.29	18.2	16.4	1.8	
	75	1.42	24.9	21.9	3.0	
	100	0.69	31.3	26.3	5.0	

Table 9: Langmuir isotherm constants for yellow 5 GL dye ions

Dye	Temperature (K)	Q_m (mg/g)	B (L/mg)
Yellow 5 GL	303	37.9	0.05
	313	39.5	0.06
	323	45.0	0.07
	333	47.8	0.10

Table 10: Freundlich isotherm constants for yellow 5 GL dye ions

Dye	Temperature (K)	n	K_f (mg/g)
Yellow 5 GL	303	1.9	3.4
	313	2.0	4.6
	323	1.9	5.4
	333	1.9	6.6

Table 11: Temkin isotherm results for the adsorption of yellow 5 GL dye ions

Dye	Temperature (K)	b_T(J/mg)	a_T(L/g)
Yellow 5 GL	303	292.1	0.4175
	313	291.8	0.5891
	323	263.9	0.6939
	333	258.6	1.0966

4. CONCLUSION

The adsorption of Yellow 5GL acid dye from aqueous solution on the adsorbent prepared from the seeds of Sugar Apple was depend on pH and maximum dye was removed at pH 3. The best fitting of experimental equilibrium data for Langmuir isotherm revealed monolayer adsorption. The kinetic study of the adsorption process was found to be pseudo-second order. The thermodynamic

parameters determined were revealed the adsorption was thermo dynamically feasible, spontaneous, endothermic and increase in the randomness at the solid - liquid interface. The results confirmed that the activated carbon prepared from the seeds of Sugar Apple is an effective and economically viable adsorbent for the removal of the dye from the aqueous solution.

CONFLICT OF INTEREST

Authors have no conflict of interest.

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