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ORIGINAL ARTICLE

Removal of Organic Contents from Wastewater Using *Leusas aspera*

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ABSTRACT
The removal of COD from wastewater has been carried out using leusas aspera as an adsorbent. The adsorption efficiency of leusas aspera leaves (LAC) stems (SAC) and roots (RAC) were investigated. The plant leusas aspera was carbonized as at 300^{0}C then is was activated in a furnace at very high temperature around 400^{0}C with steam in absence of air. The experimental batch equilibrium data was correlated by Freundlich and Langmuir isotherms. The adsorption data fitted well in to the Freundlich isotherm. The organic concentration expressed as COD was reduced from initial value of 214 ppm in to 188.8 ppm by using SAC. 201.6 ppm by using the adsorbent of LAC and 192 ppm by using the adsorbent of RAC. The removal efficiency increases as adsorbent dose increases. The overall result shows that leusas aspera activated carbon can be fruitfully used for the removal of COD from polluted water.
KEY WORDS: *Leusas Aspera, LAC, RAC, SAC, Adsorption, Freundlich isotherms and Langmuir isotherms.*

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INTRODUCTION

The discharge of industrial effluents into surface water is the serious environmental problem. Wastewater effluents contain large amount of organic and inorganic chemicals having stable compounds like Nitrate, Phosphate, Chloride, Sulphate etc., which their removal from polluted source is difficult. The removal of pollutant from aqueous waste stream by adsorption using AC in fixed beds is an important industrial waste water treatment process [1]. Activated carbon was used to remove highly odorous dissolved organic compounds from industrial waste water [2]. AC produced from jack fruit peel, (carbonized by chemical method) to treat and remove malachite green from wastewater obtained from a dye industry [3]. For phenol, the former exhibited a slightly higher adsorption than the latter. The use of locally prepared AC from palm date use of locally prepared activated carbon from palm date pits results showed that it is more efficient than the commercial sample. The phenol removal efficiency was investigated at several pH values, carbon dosages and contact times. Freundlich adsorption isotherm was used to analyze the adsorption efficiencies of activated carbon is the most widely used adsorbent in the treatment of wastewater, due to its high adsorption capacity and rate. Activated carbon are prepared and used in different shapes including powered activated carbon and granular activated carbon and activated carbon fiber. Activate carbon with surface area of 110.35 – 146.06 m^2g^{-1} from sunflower seed were applied to removal of acid blue [4]. Among them adsorption process found to be the most effective method. Adsorption as a water treatment process has aroused considerable interest during recent years.

MATERIALS AND METHODS

The material used for the experiment were activated carbon produced from leaves (LAC), stems (SAC) and roots (RAC) of *leusas aspera*. Water samples were collected directly from bore wells in my study area located nearby industrial site of Chennai city (south India) by using clean labeled stoppered polythene bottles and it was preserved in a refrigerator. The analyze were carried out according to ISI standard methods [5]. The pH and TDS were measured by electrometric method by using digital pH meter-E1-model (111E) and TDS meter labtronics–LT15 model. Colour was measured by platinum-cobalt method. Nitrate was measure spectrophotometric method by using spectrophotometer-deep vision model-305E. COD was determined by open reflux method. Analytical grade chemicals were used throughout the study without further purification. To prepare all the reagents and calibration standards, double distilled water was used.

ACTIVATED CARBON

The stems, roots and leaves of *leusas aspera* collected from local area of Thiruvallur district and washed with double distilled water. It was dried in sunlight; the material is heated until to create a char, this char is than activated in a furnace at very high temperature around 300 to 400^{0}C with steam in the absence of air. The carbonized leusas is cooled off, the soluble ash content was removed by using double distilled water and dried in an oven at 110^{0}C for 24 hrs.The final product was kept in an air tight polyethylene bag, the physical properties were analyzed by using Scanning Electron Microscope (SEM).

ADSORPTION OF ORGANIC CONTAMINANTS

The adsorption equilibrium studies were carried out by using granular activated carbon produced of mass 3, 5, 7 and 9 g. Each mass was placed in a 250 ml Erlenmeyer flask contains 100 ml of wastewater sample. The flask contains 9 g carbon was shaken continuously while the flask contains 3, 5 and 7 g of activated carbon were shaken intermittently for 3 hrs. The activated carbon samples were percolated with the wastewater effluent until equilibrium was achieved and this occurred within 3 hrs of shaking. Samples were taken at five minutes interval and the resulting mixture from each flask was filtered and the COD of the filtrate was determined. The amount of organic removal achieved at the varying dosages of carbon gave an indication of activated carbon usage rate required to treat wastewater to a specified effluent quality, as well as the type of solid-fluid phase equilibrium that exists for the particular case under consideration. The equilibrium data obtained were processed to understand the adsorption of the contaminant on to the activated carbon produced using the equilibrium isotherm equation of Freundlich and Langmuir which are commonly applied in water and waste water treatment.

DETERMINATION OF COD

To determine the COD, 25 ml of 0.02 N standard potassium dichromate solutions was added to 40 ml of waste water sample in a 250 ml round bottom flask. 10ml of acid-silver reagent were added. A few glass beads were added to serve as anti-bumping aid, and the flask was connected to reflux condenser. Turn on cooling water add additional 70 ml of sulphuric acid reagent through open end of condenser, with swirling and mixing. Reflux for 2 hours; cool, wash down condenser with distilled water to double the volume of contents, cool. Add 2 drops of Ferroin indicator; titrate with standardized FAS solution until there was a change of colour from bluish green to reddish brown. The blank value was determined in the same way with 40ml of distilled water. The COD Values of the respective samples were calculated using following equation.

$$CODmgO/l = \frac{(A - B)\times M \times 8000}{ml.of.sample}$$

A = FAS used for blank, mL
B = FAS used for sample, mL
M = Molarity of FAS

RESULT AND DISCUSSION

ACTIVATED CARBON CHARACTERIZATION

The surface characteristics of activated carbon prepared from leaves (LAC), stems (SAC) and roots (RAC) of *leusas aspera* (bulk density, porosity, pore volume, ash content pH, average particle size, iodine number etc.,) are given in table 1. SEM micrographs of the prepared samples are shown in Fig. 1, 2 and 3 shows the porosity of AC from leaves, stems and roots of *leusas aspera* respectively.

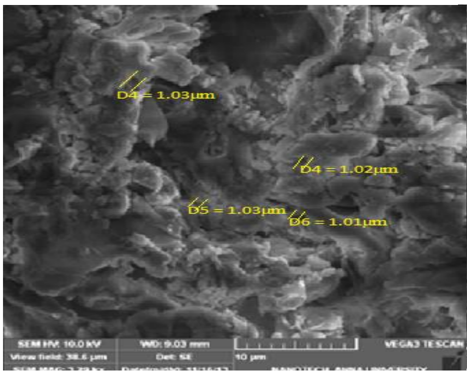

Figure-1 SEM of root

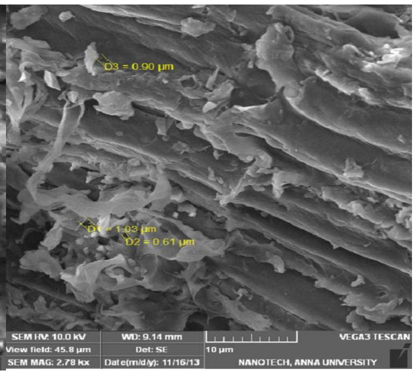

Figure-2 SEM of stem

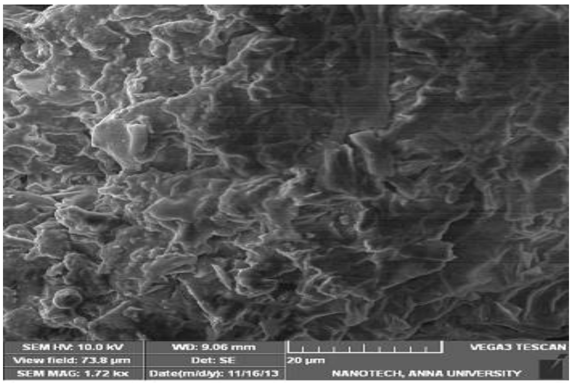

Figure-3 SEM of leaves

Table -1Characteristics of Activated carbon of leusas aspera and commercial activated carbon

Parameter	Leaves of leusas aspera (LAC)	Stems of leusas aspera (SAC)	Roots of leusas aspera (RAC)	Commercial activated carbon
Bulk Density	0.5	0.5	0.5	0.4
Porosity	0.19	0.18	0.206	0.214
Pore volume	0.95µm	0.90µm	1.03µm	1.109 µm
Ash content	18	14	8	8
Average particle size	27.63	29.66	27.56	5-50

Iodine number and pH of the activated carbon were 1098, 1022, 1116 mg g-1 and 6.3, 6.7 and 6.9 respectively. Table 2 shows the amount of pollutants present in groundwater sample obtained from my study area located near by the industrial site. The sample A, B and C were collected at different polluted site in Chennai city. A was collected from Thiruvangadam Nagar (west Chennai), Sample B was collected from V.M.Street (central Chennai) and sample C was collected from P.Koil Street (south Chennai). The physical and chemical analyses for these samples were carried out using Indian standard method. These three samples have higher concentration of various physico-chemical parameters.

Table -2 various physico-chemical parameters of water sample

Parameters	Sample A	Sample B	Sample C
pH	6.82	7.99	7.89
TDS (in ppm)	1486	1456	974
COD (in ppm)	214	218	164
Nitrate (in ppm)	42	18	18
Colour	2TCU	2TCU	2TCU

EFFECT OF pH

Adsorption of COD was studied over the pH range 2.0 - 9.0 (Fig. 4). Experiments were Carried out at 214 mg L-1 initial COD concentration with 3g adsorbent mass at room temperature (39 ± 1^{o}C) for 3 hour equilibrium time. The maximum Adsorption was observed at pH 4.0 for LAC, SAC and RAC. At Only acidic pH (pH = 4) the adsorbent surface get favors uptake of COD.

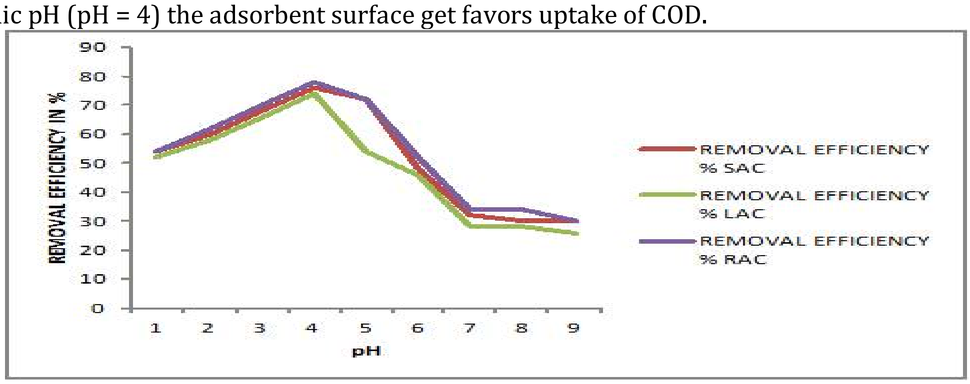

Figure-4 Effect of pH on COD Removal.

EFFECT OF CONTACT TIME

The rate of adsorption is important for designing batch adsorption experiments. The adsorption studies were carried out at fixed adsorbent dose (3g), room temperature (39 ± 1°C), pH (7.0) and at different initial concentrations of COD (218,214 and 164 mg L-1) for different time intervals (15, 30, 45, 60, 90 and 120 min).The results are shown in Table 3.

Table -3 Effect of contact time on adsorption for COD by SAC, LAC and RAC
(dosage = 3 g/100 mL, pH = 4, temp = 300 K)

Initial COD concentration (mg/l)	% of COD removal with time (min) SAC					
	15	30	45	60	90	120
214	5.2	6.8	8.3	11.2	11.4	11.5
218	6.2	7.2	8.8	10.8	11.6	11.6
164	5.8	6.8	8.9	10.8	11.4	11.4
	% of COD removal with time (min) LAC					
214	3.2	4.4	5.2	6.4	6.5	6.8
218	3.8	4.6	5.2	6.2	6.4	6.5
164	3.8	4.4	5.3	6.2	6.5	6.7
	% of COD removal with time (min) RAC					
214	5.2	6.4	8.4	11	11.2	11.3
218	5.8	6.8	8.2	10.8	11	11.2
164	5.8	6.6	8.9	11	11.3	11.4

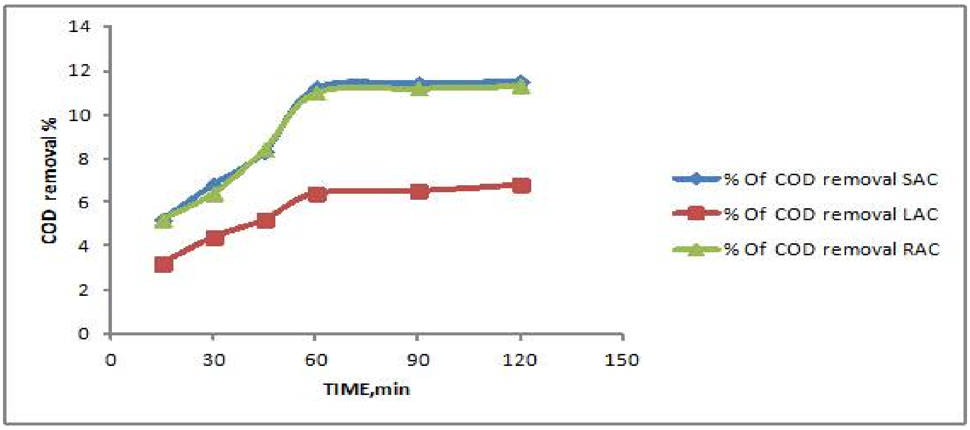

Figure-.5. Effect of contact time on adsorption for COD by SAC, LAC and RAC

The adsorption efficiency of COD increased considerably until the contact time reached 60 min**.** Further increase in contact time did not enhance the adsorption.

EFFECT OF ADSORBENT DOSAGE

The experimental batch adsorption data obtained for sample A, B and C are presented in table 4. These studies were carried out at different initial concentration of COD water sample for varying the dosage of adsorbent (3, 5, 7 and 9 g). The increase in adsorbent dosage caused decrese in the percentage of COD removal. It is clear that the removal of COD in wastewater sample depends on the concentration of adsorbent of stems (SAC), roots (RAC) and leaves (LAC) of *leusas aspera*. The results show that the adsorbent has good potential of COD removal. The batch adsorption experiment carried out to establish the nature of equilibrium that existed in the COD water activated charcoal derived from *leusas aspera* indicated that 3 g of SAC could remove 6, 6.8 and 7.2 mg of total COD content of wastewater sample (i.e., A, B, and C). It can be observed from the equilibrium concentration x / m decrease with increase in carbon dosage and also these results indicated the concentration of organic pollutant decreases with increase in carbon dosage of SAC, RAC and also these results indicates excessive percentage of COD removed by adding adsorbent dosage of SAC and RAC. If LAC having to remove only less % of organic pollutant.

Table-4 Expremental batch adsorption data for sample A, B and C

Initial COD concentration (mg/l)	COD removal with different adsorbent dosage of stems (g/l)			
	3g	5g	7g	9g
214	6	12.4	22	25.2
218	6.8	13.2	19.6	26
164	7.2	10.4	16.8	23.2
	COD removal with different adsorbent dosage of Leaves (g/l)			
214	2.8	6	9.2	12.4
218	3.6	6.8	13.2	16.4
164	4	7.2	10.4	16.8
	COD removal with different adsorbent dosage of roots (g/l)			
214	6	9.2	15.6	22
218	6.8	10	16.4	22.8
164	7.2	10.4	13.6	20

ADSORPTION ISOTHERM MODELING

The experimental data were analyzed to the near form of Langmuir and Freundlich isotherms. The Freundlich isotherm is represented by the following equation.

$$\log \frac{x}{m} = \log k + \frac{1}{n} \log c$$

Here, k and n are constants incorporating all factors affecting the adsorption capacity and intensity of adsorption respectively. Linear plot of log x/m versus log c shows that the adsorption of COD follows the Freundlich adsorption isotherm (fig.6, 7 and 8) the values of k and n were calculated from the intercept and slope of the plot. The magnitude of the exponent n gives the indication of favorability and k the capacity of adsorbent/adsorbate system. The n values in between 1 and 10 representing beneficial adsorption [6 -7].

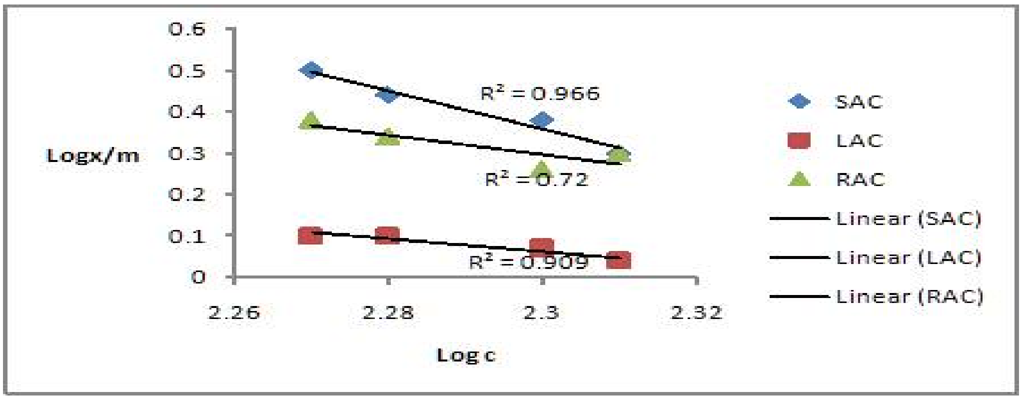

Figure-6. Comparison of Freundlich isotherms of sample A.

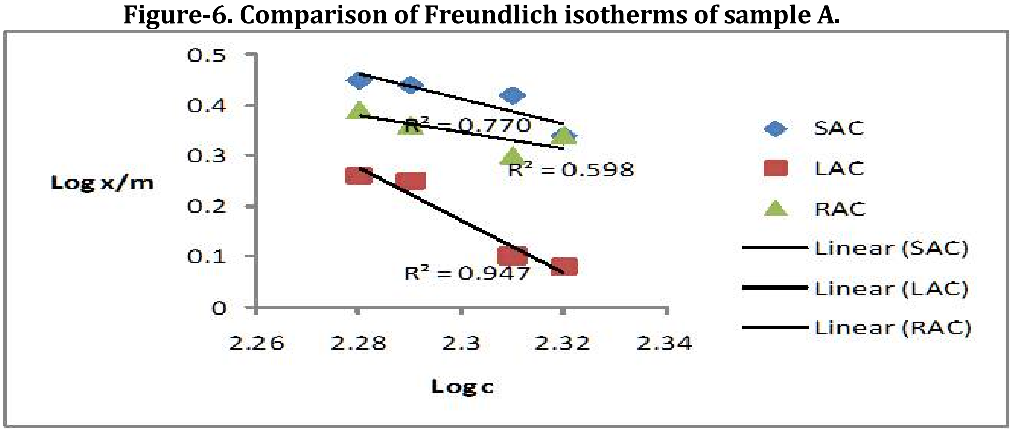

Figure-7. Comparison of Freundlich isotherms of sample B.

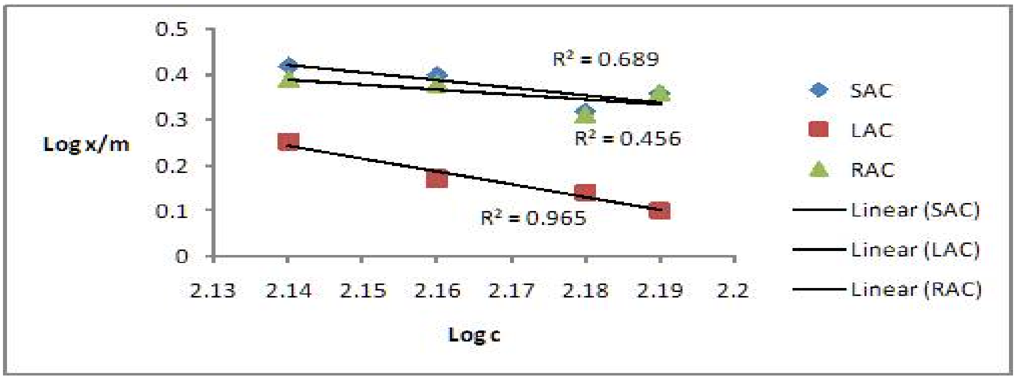

Figure-8. Comparison of Freundlich isotherms of sample C.

The Langmuir isotherm's represented by the following equation

$$\left(\frac{Ce}{qe}\right)=\left(\frac{1}{Qob}\right)+\left(\frac{Ce}{Qo}\right)$$

Here Ce (mg/l) equilibrium concentration of COD, qe is the amount of COD adsorbed at equilibrium (mg/l). Qo and b is Langmuir constants related to the adsorption capacity and energy of adsorption respectively. The linear plot of Ce/qe versus Ce suggests the applicability of the Langmuir isotherms (fig.9, 10, and 11). The values of Qo and b were determined from the slope and intercept of the plot.

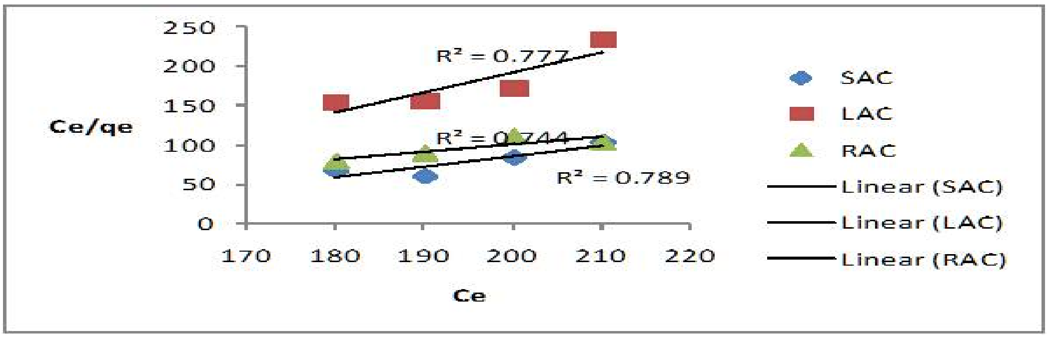

Figure-9. Comparison of Langmuir isotherms of sample A.

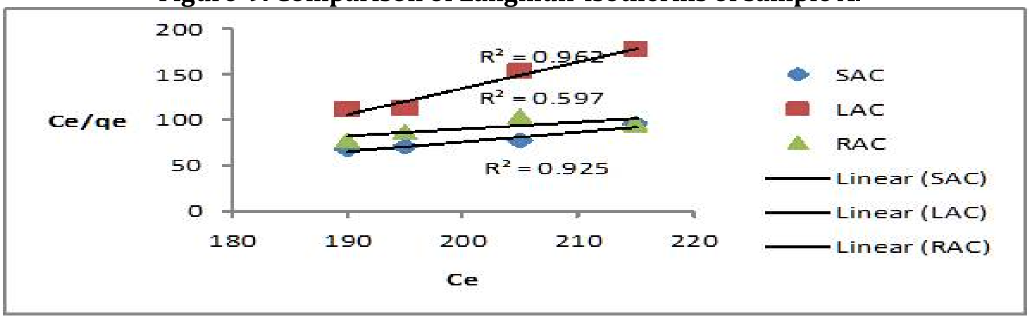

Figure-10. Comparison of Langmuir isotherms of sample B.

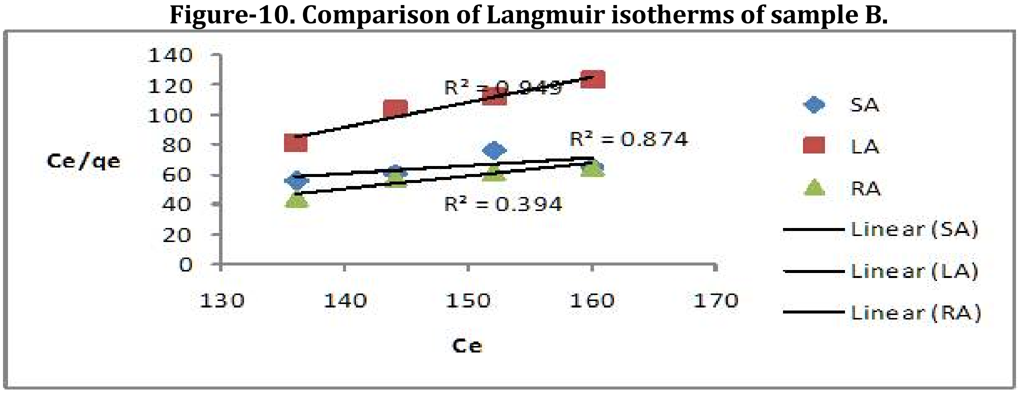

Figure-11. Comparison of Langmuir isotherms of sample C.

Table-5 Adsorption isotherm constants and coefficient of determination of different adsorbent

Sample	Adsorbent	Freundlich isotherm constants			Langmuir isotherm constants		
		K (mg/g)	1/n	R^2	Q_o	R_L	R^2
A	SA	10.93	4.6	0.96	1.35	21×10^{-4}	0.78
A	LA	3.51	1.5	0.90	2.53	14×10^{-4}	0.77
A	RA	5.51	2.4	0.72	0.93	54×10^{-4}	0.74
B	SA	5.93	2.4	0.77	1.09	32×10^{-4}	0.92
B	LA	4.02	1.6	0.94	2.88	10×10^{-4}	0.96
B	RA	11.94	5.2	0.59	0.78	69×10^{-4}	0.54
C	SA	3.97	1.66	0.68	0.53	44×10^{-3}	0.34
C	LA	6.26	2.81	0.96	1.68	42×10^{-4}	0.94
C	RA	2.71	1.08	0.45	1.23	78×10^{-4}	0.39

The essential features of Langmuir isotherm can be expressed in terms of dimensionless constant separation factor, R_L that is given as follows.

$$R_L = \frac{1}{(1+bCo)}$$

Where Co is initial concentration of adsorbent and b is Longmuir constant. The value of R_L indicate the type of isotherm to be either favorable ($0 < R_L < 1$), unfavorable ($R_L > 1$), Linear ($R_L = 0$). The value of R_L was found to be ($0 < R_L$) are given in table 5. This value suggesting that the isotherm to be favorable at the concentration studied. According to R^2 values, Freundlich model better describes the equilibrium adsorption. These coefficients of determination and the isotherm constant are given in (table 5). The high value of R^2 for the two isotherms indicates that the adsorption of COD could be well described. Hence Freundlich isotherm assume that adsorption site have different affinities for different adsorbate species [8]. Thus it is better describes the adsorption processes in real aquatic environment. According to 1/n is a measure of adsorption intensity. A value of 1/n<1 shows a normal adsorption. while 1/n >1 is indicate of cooperative adsorption, [9-10] the value of 1/n was found to be is greater than 1 and also these value exceed by using the adsorbent like SA and RA of leusas aspera are given in (table 5).

Table-6 Various physico-chemical parameters of water sample A.B and C after adsorption treatment.

Samples	pH	TDS (ppm)	Nitrate (ppm)
A (after SAC Treatment)	6.92	1414	18
A (after LAC Treatment)	6.87	1436	26
A (after RAC Treatment)	6.91	1412	22
B(after SAC Treatment)	8.07	1388	8
B (after LAC Treatment)	8.05	1414	12
B (after RAC Treatment)	8.09	1391	6
C (after SAC Treatment)	7.96	906	6
C (after LAC Treatment)	7.93	928	14
C (after RAC Treatment)	7.95	902	8

Comparing the result obtained from table 2. With the sample specification on table.6 it can be observed that pH value was slightly increases (The range up to 0.14). The results obtained showed that there was a remarkable decrease in the amount of TDS after the adsorption. It was reduced from 70-75 ppm by using the adsorbent like SAC and RAC. If 45-53 ppm of TDS was removed by using the adsorbent of LAC. Similarly nitrate concentration is also reduced from 20-24 ppm.

CONCLUSION

This present study has revealed some concealed facts about the usefulness and effectiveness of granular activated carbon produced from stems (SAC), roots (RAC) and leaves (LAC) of *leusas aspera*. This activated carbon can effectively removed COD from wastewater. The adsorption of COD was depending on the adsorbent dosage and also the adsorption obeyed both Langmuir and Freundlich isotherms. The equilibrium data for the adsorption fitted better in to the Freundlich equation. The organic concentration expressed as COD was reduced from initial value of 214,218 and 164 ppm in to 188.8, 192.8 and140.8 ppm by using the adsorbent of SAC. 201.6, 201.6, and 147.2 ppm using the adsorbent of LAC and

192,195.2 and 144 ppm by using the adsorbent of RAC. Hence significant pollutant removal efficiency rates of SAC 11%, LAC 6% and RAC 10% were achieved by using little quantity of adsorbent. Therefore the effectiveness of the activated carbon produced from *leusas aspera* in the removal of organic contaminant has been established. The study also proved that natural adsorbent such as activated carbon of leusas aspera is alternative option for COD removal from wastewater. Works continue on the effect of particle size, fixed bed, and flow time on the efficiency of the treatment.

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