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ORIGINAL ARTICLE

Adsorption of chromotrope dye onto activated carbons obtained from the seeds of various plants: Equilibrium and kinetics studies

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Tamarind seed carbon (TSC);
Soapnut carbon (SNC);
Commercial activated carbon (CAC)

Abstract The use of low-cost, easily obtained and eco-friendly adsorbents has been employed as an ideal alternative to the current expensive methods of removing dyes from wastewater. This study investigates the potential use of activated carbons such as aamla seed carbon (ASC), jambul seed carbon (JSC), tamarind seed carbon (TSC) and soapnut carbon (SNC) for the removal of chromotrope (CH) dye from simulated wastewater. The obtained results were compared with that of Commercial Activated Carbon (CAC). The effects of different system variables, adsorbent dosage, pH and particle size were studied. The results showed that as the amount of the adsorbent increased, the amount adsorbed per unit mass decreased accordingly. Optimum pH value for dye adsorption was determined as ~2.0. The adsorption of chromotrope fit well into pseudo-second-order rate equation. The adsorption also obeyed Langmuir and Redlich–Peterson equations better than Temkin, Freundlich and Dubinin–Radushkevich (D–R) equations. The adsorption capacities of low cost adsorbents were found to be in the following order ASC > JSC > TSC > SNC. The activated carbons prepared were characterized by FT-IR and SEM analysis.

1. Introduction

Colour is the most obvious indicator of water pollution. The discharge of coloured wastes into streams not only affects their aesthetic nature but also interferes with the transmission of sunlight into streams and therefore reduces photosynthetic action. Effluents from the dye manufacturing industry, textile industry, and pulp and paper industry are highly coloured.

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The dye under investigation, chromotrope (CH) is a red azo dye. Azo dyes constitute the largest and most versatile class of synthetic dyes used in the textile, pharmaceutical, food and cosmetic industries. They are characterized by nitrogen to nitrogen double bonds and are a major source of aromatic amines having in their molecular structure one or more aromatic rings with one or more amino substituents. The colour of the dyes is due to the azo bond and associated chromophores. The biological reduction of an azo bond is responsible for the appearance of dangerous aromatic amines in liquid effluents, such as aniline and sulphanilic acid which are carcinogenic. Strong colour imparted by the dyes poses aesthetic problems and serious ecological problems such as inhibition of benthic photosynthesis and carcinogenicity. In some cases they can be harmful compounds and can originate dangerous compounds through oxidation, hydrolysis and other chemical reactions, being thus of particular environmental concern (Puvaneswari et al., 2006).

Considerable research has been done on colour removal from these wastes. Many treatment processes have been applied for the removal of dyes from wastewater such as: photocatalytic degradation (Sohrabi and Ghavami, 2008), sonochemical degradation (Abbasi and Asl, 2008), micellar enhanced ultrafiltration (Zaghbani et al., 2008), cation exchange membranes (Wu et al., 2008), electrochemical degradation (Fan et al., 2008), adsorption/precipitation processes (Zhu et al., 2007), integrated chemical–biological degradation (Sudarjanto et al., 2006), solar photo-Fenton and biological processes (Garcia-Montano et al., 2008), Fenton-biological treatment scheme (Lodha and Chaudhari, 2007) and adsorption on activated carbon (Nevine Kamal Amin, 2008; Gupta et al., 2006). As synthetic dyes in wastewater cannot be efficiently decolorized by traditional methods, the adsorption of synthetic dyes on inexpensive and efficient solid supports was considered as a simple and economical method for their removal from water and wastewater. The adsorption process provides an attractive treatment, especially if the adsorbent is inexpensive and readily available. In a country like India, where 80% of the population depends on the land for a living, the utilization of agricultural waste is of great significance. In India, more than 200 million tons of agricultural residues are generated annually.

In recent years, special emphasis on the preparation of activated carbons from several agricultural by-products has been given due to the growing interest in low cost activated carbons for application concerning treatment of wastewater. Researchers have studied the production of activated carbon from palm-tree cobs (Avom et al., 1997), plum kernels (Wu et al., 1999), cassava peel (Rajeshwarisivaraj et al., 2001), bagasse (Tsai et al., 2001), jute fibre (Senthilkumar et al., 2005), coconut husk (Tan et al., 2008), rattan sawdust (Hameed et al., 2007), seed shells (Thinakaran et al., 2008), cocoa shell (Theivarasu et al., 2011) and Eucalyptus globules Bark (Kannan and Pagutharivalan, 2012). The advantage of using agricultural by-products as raw materials for manufacturing activated carbon is that these raw materials are renewable and potentially less expensive to produce.

The objective of this present study is to explore the synthesis and feasibility of using activated carbons ASC, JSC, TSC and SNC as adsorbents for the removal of chromotrope, as an alternative to CAC which is most widely used in various textile-processing industries.

2. Experimental

2.1. Preparation of activated carbons

The material commercial activated carbon (CAC) was supplied by S.D. Fine Chem Ltd., India. Raw materials for the preparation of low cost carbons such as Indian Gooseberry or aamla (Phyllanthus Emblica) Seed, Jambul (Syzygium cumini) Seed, Tamarind (Tamarind Indicus) seed and Soapnut (acacia sinuata) seed were selected because of their easy availability, abundance, cheap market price to provide an economical method of waste management. The agricultural by-products or wastes were collected locally from market, washed and cleaned with water and air dried. These raw materials were cut into small pieces and then carbonized in the presence of $NaHCO_3$, pyrolysed and activated by acid digestion with 4 N HNO_3. The pyrolysis method is a process consisting of dehydration and carbonization of the raw materials at low temperature (300–400 °C) followed by activation in the absence of air by CO_2 or steam at high temperature (700–900 °C). Carbonization was carried out by pyrolysis of the raw materials such as Aamla seed (AS), Jambul seed (JS), Tamarind Seed (TS) and Soapnut seed (SN). The raw materials were locally collected, cut into small pieces, washed with water, dried and carbonized at the temperature range of 300–400 °C, in a muffle furnace (Neolab, AUS-101, India) in the absence of air. During carbonization, volatile products were removed. The residue remaining after pyrolysis is referred to as char. After the carbonization and steam activation the carbons were collected, washed, dried and stored in brown bottles. Before carrying out each adsorption experiment the carbons were activated and used.

2.2. Adsorbate

The dye chromotrope (Fig. 1) used in this work was purchased from BDH (India). The CH (C.I.14720) used in this study has a molecular weight of 502.43 g/mol, with a molecular formula of $C_{20}H_{12}N_2S_2O_7Na_2$. The maximum wavelength of absorption of this dye is 515 nm.

2.3. Equilibrium analysis

The adsorption experiments were carried out in a batch process (Robinson et al., 2001). Adsorption experiments were carried out by adding a fixed amount of adsorbent (2 g L^{-1} for CAC and 10 g L^{-1} for other carbons) into a number of 250 mL-stoppered flasks containing a definite volume (50 ml in each case) of different initial concentrations (8–225 mg/L) of dye solution without changing pH and temperature 30 °C. The flasks were placed in a thermostatic water-bath shaker

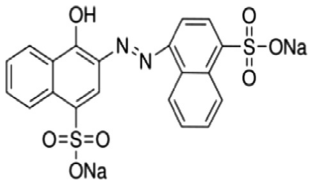

Figure 1 Structure of chromotrope.

and agitation was provided at 130 rpm for 30 min to ensure equilibrium was reached. At time $t = 0$ and equilibrium, the absorbance of dye solution was determined by the ELICO microprocessor photo colorimeter, at $\lambda_{\max}$ (515 nm) value of dye. The amount of adsorption at equilibrium, qe (mg/g), was calculated by

$$\text{Amount adsorbed } (q_e) = (C_i - C_e)/m \tag{1}$$

where, C_i and C_e were the initial and equilibrium concentrations (in mgl^{-1} or ppm) of dye, respectively and m is the weight of AC (in gl^{-1}). The dye removal percentage can be calculated as follows:

$$\text{Percentage removal} = 100(C_i - C_e)/C_i \tag{2}$$

The effect of dose of adsorbents on the amount of CH adsorbed was obtained by adding different amounts of adsorbents (1.4–2.6 g l^{-1} for CAC and 8–16 g l^{-1} for other carbons) into solutions of fixed initial concentration (8–25 ppm) of dye solution without changing pH (5.0), temperature 30 °C, particle size (90 microns) and contact time (30 min). The effect of pH was also observed by varying the pH of the solution, keeping all others constant. Adsorption studies of dyes were carried out at different particle sizes of adsorbent with various indigenously prepared ACs (range: 90–250 microns) except CAC, under constant optimum initial concentration, contact time (Table 1) optimum dose of adsorbent and pH of the dye solution at 30 °C. The values of percentage removal and amount of dye adsorbed (in mg g^{-1}) are calculated and plotted against the particle size of the adsorbents. In order to study the kinetics/dynamics of adsorption of dyes, the adsorption experiments were conducted by varying the contact time (range: 5–60 min.) at fixed optimum initial concentration of dyes with a fixed dose of adsorbent (CAC = 1 g l^{-1} and ACs = 10 g l^{-1}) and particle size (90 microns) at 30 °C and at the pH of the solution. The dye concentrations were measured at different time intervals.

2.4. Instrumental study

The large flakes or coarse particles of the indigenously prepared adsorbents (*viz*., ASC, JSC, TSC and SNC) were grounded and sieved in a mechanical siever (Jayanth brand, India) to different consistent discrete particle sizes. The pH measurement was done by using Systronics digital pH metre (model: 335). The FT-IR spectra of the adsorbent materials (ACs), before and after adsorption of CH were recorded in KBr pellets by using a BIO-RAD WIN IR Spectrometer (Frequency range: 400–4000 cm^{-1}). The scanning electron microscope (SEM) photographs of the adsorbent materials (ACs), before and after adsorption of CH dye were obtained using a JEOL JSM – 5300 SEM Instrument.

3. Results and discussion

3.1. Adsorption isotherm studies

Several mathematical models can be used to describe experimental data of adsorption isotherms. In this work, the equilibrium data for CH on CAC, ASC, JSC, TSC and SNC were modelled with the Langmuir, Freundlich, Temkin, Redlich – Peterson Isotherm and Dubinin–Radushkevich isotherm models.

Table 1 Experimental conditions for the adsorption of chromotrope on CAC, ASC, JSC, TSC and SNC.

No.	Variation in chromotrope	Initial concentration (ppm)		Contact time (min.)		Dose of adsorbent (gL^{-1})		Initial pH		Particle size (micron)		Temperature (°C)	
		CAC	ASC JSC TSC SNC	CAC	ASC JSC TSC SNC	CAC	ASC JSC TSC SNC	CAC	ASC JSC TSC SNC	CAC	ASC JSC TSC SNC	CAC	ASC JSC TSC SNC
1	Initial concentration	100–225	10–25 10–25 8–16 8–16	30	30	2	10	5.0	5.0	90	90	30	30
2	Contact time	125	10 10 8 8	5–60	5–60	2	10	5.0	5.0	90	90	30	30
3	Dose of adsorbent (g L^{-1})	125	10 10 8 8	30	30	1.4–2.6	8–10	5.0	5.0	90	90	30	30
4	Initial pH	125	10 10 8 8	30	30	2	10	2–11	2–11	90	90	30	30
5	Particle size	–	10 10 8 8	–	30	–	10	–	5.0	–	90–250	–	30

Table 2 Results of correlation analysis on testing the applicability of adsorption isotherm for the removal of chromotrope dye by adsorption on CAC, ASC, JSC, TSC and SNC.

S. No.	Parameters	Adsorbents				
		CAC	ASC	JSC	TSC	SNC
I	*Freundlich isotherm*					
	Slope ($1/n$)	0.4	0.5625	0.5216	0.309	0.3517
	Intercept ($\log k$)	0.09	0.23	−0.1082	−0.034	−0.1179
	r-Value	0.985	0.953	0.999	0.975	0.974
II	*Langmuir isotherm*					
	Slope ($1/Q_0$)	0.0526	0.0839	0.0856	0.1997	0.2057
	Intercept ($1/Q_0$b)	0.024	0.6378	1.039	0.6559	0.8517
	r-Value	0.976	0.999	0.991	0.997	0.995
	Q_0 (mg g^{-1})	190	11.91	11.68	5.0	4.86
	b (g L^{-1})	0.2193	0.1317	0.0823	0.3044	0.2416
	$R_L = 1/(1 + C_0b)$	0.0352	0.0002	0.0002	0.0002	0.0002
III	*Temkin isotherm*					
	A(L/mg)	1.159	1.034	0.669	3.452	2.236
	B	52.41	2.885	2.79	1.06	1.105
	r-Value	0.962	0.999	0.991	0.978	0.977
IV	*Redlich–Peterson isotherm*					
	K_R	41.67	1.47	0.962	1.525	1.174
	αR	0.578	1.018	0.8429	0.6058	0.5511
	β	0.8922	1.049	1.079	0.9834	0.9974
	r-Value	0.989	0.999	0.991	0.997	0.995
V	*Dubinin–Radushkevich isotherm*					
	X'_m (mg g^{-1})	166.7	8.355	7.035	4.143	3.877
	E (kJ mol^{-1})	445.3	415.1	356.2	561.4	504
	K' (mol^2 kJ^{-2}) 10^{-6}	−2.5	−2.9	−3.9	−1.6	−2.0
	r-Value	0.866	0.987	0.954	0.984	0.986

The coefficient of correlation (*r-value*) obtained indicates that Langmuir and Redlich – Peterson Isotherms provided a better fit for the adsorption of CH on all the carbons. The values of the Langmuir constants obtained in this study are presented in Table 2. Langmuir parameters have been obtained from the linear correlation between the values of (C_e/q_e) and C_e (Fig. 2a). The essential characteristics of the Langmuir isotherm can be expressed in terms of a dimensionless constant separation factor R_L that is given by the following equation:

$$R_L = 1/(1 + bC_i) \quad (3)$$

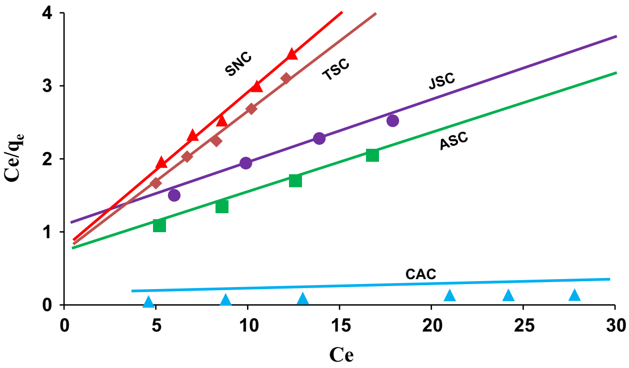

Figure 2 (a) Langmuir adsorption isotherm of chromotrope on various carbons. (b) Redlich–Peterson adsorption isotherm of chromotrope on various carbons.

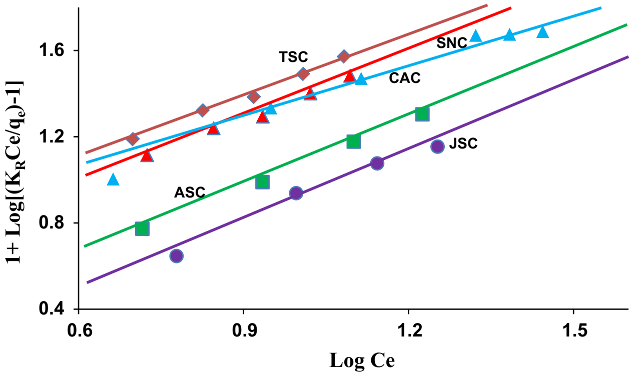

Fig. 2 (*continued*)

where, b is the Langmuir constant (in L mg^{-1}) and C_i is the (optimum) initial concentration (in mg L^{-1}) in the adsorption studies. The value of R_L infers the nature of adsorption isotherm and the feasibility of adsorption process. The value of R_L indicates the shape of the isotherm to be either unfavourable ($R_L > 1$), linear ($R_L = 1$), favourable ($0 < R_L < 1$), or irreversible ($R_L = 0$). The R_L value for the adsorption of CH onto various carbons is in the range of [0.0002–0.0352], indicating favourable adsorption. Similar report was given by Dulman et al. for the removal of some textile dyes by beech wood sawdust (Dulman and Cucu-Man, 2009). The applicability of the Langmuir isotherm suggests the mono-layer coverage of the chromotrope dye on the surface of the carbon. The

Table 3 Kinetics and dynamics of adsorption of chromotrope dye on CAC, ASC, JSC, TSC and SNC.

S. No.	Parameters	Adsorbents				
		CAC	ASC	JSC	TSC	SNC
I	*First order equation*					
	Correlation coefficient (r)	0.818	0.889	0.932	0.985	0.952
	k (min^{-1})	44.8	2.95	9.76	16.68	16.21
II	*Natarajan and khalaf equation*					
	Correlation coefficient (r)	0.962	0.941	0.937	0.932	0.954
	k (min^{-1})	0.1389	0.0254	0.0122	0.182	0.0151
III	*Bhattacharya and venkobachar equation*					
	Correlation coefficient (r)	0.995	0.971	0.987	0.997	0.990
	k (min^{-1})	0.1864	0.0959	0.0694	0.0563	0.0548
IV	*Pseudo first order equation*					
	Correlation coefficient (r)	0.996	0.912	0.987	0.997	0.989
	k (min^{-1})	0.1864	0.068	0.0694	0.0527	0.0549
V	*Pseudo second order equation*					
	Correlation coefficient (r)	0.999	0.980	0.989	0.992	0.976
	k (min^{-1})	7.003	0.2232	0.1648	0.1223	0.106

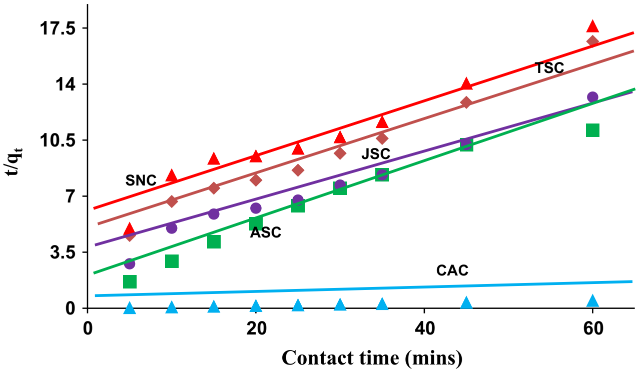

Figure 3 Pseudo second order rate equation for the adsorption of chromotrope on various carbons.

mono-layer adsorption capacities (Q_o) of the adsorbents are found to be of the order given below:

SNC < TSC < JSC < ASC << CAC

Among the low cost carbons, ASC was found to have the highest value of Q_0 (11.91) as shown in Table 2. The Redlich–Peterson isotherm (R–P) is a combination of features of both Langmuir and Freundlich isotherms. It can be used to describe adsorption on both homogenous and heterogeneous surfaces. The linearized form of R–P isotherm can be given as:

$$\log[(K_R C_e/q_e) - 1] = \beta \log C_e + \log(\alpha R) \tag{4}$$

By plotting the values of log [($K_R C_e/q_e$)−1] against log C_e, the values of (αR) and β can be determined from the intercept [log (αR)] and the slope (β), respectively (Fig. 2b). Except for CAC, all other carbons have r-value equal to 0.99, indicates the applicability of Langmuir and Redlich–Peterson isotherms.

3.2. Adsorption kinetics

The kinetics and dynamics of adsorption of chromotrope on various adsorbents have been studied by applying the various kinetic equations:

First order equation:

$$(1/q_t) = (k/q_{\max})(1/t) + (1/q_{max}) \tag{5}$$

Natarajan and Khalaf equation:

$$\log(C_i/C_t) = (k/2.303)t \tag{6}$$

Bhattacharya and Venkobachar equation:

$$\log[1 - U(t)] = -(k_a d/2.303)t \tag{7}$$

Pseudo first order model:

$$\log(q_e - q_t) = \log q_e - [k_{ad}/2.303]t \tag{8}$$

Pseudo second order model:

Table 4 Comparison of calculated and experimental q_e values and Δq values of chromotrope with CAC ASC, JSC, TSC and SNC.

Adsorbents	q_e exp. (mg g^{-1})	Pseudo first order kinetics		Pseudo second order kinetics		Δq (%)	
		q_e cal. (mg g^{-1})	R^2 value	q_e cal. (mg g^{-1})	R^2 value	Pseudo first order	Pseudo second order
CAC	120.2	16.12	0.880	121.6	0.999	106.6	3.1
ASC	5.4	2.52	0.987	5.6	0.999	88.4	13.2
JSC	4.6	5.3	0.974	5.8	0.999	95.8	10.2
TSC	3.6	3.94	0.995	4.9	0.999	88.5	6.4
SNC	3.4	4.2	0.979	5.0	0.999	87.8	10.4

Table 5 Statistical results of intra-particle models for the adsorption of chromotrope dye on CAC, ASC, JSC, TSC and SNC.

S. No.	Parameters	Adsorbents				
		CAC	ASC	JSC	TSC	SNC
I	*Intra- particle diffusion model*					
	k_p	1.8815	0.8222	0.6815	0.5083	0.495
	Correlation coefficient (*r*)	0.987	0.964	0.966	0.974	0.976
	Intercept	106.07	2.09	0.5093	0.0817	−0.149
	Δ*q* (%)	0.0022	0.1553	0.7289	0.7523	0.8911
II	*Log (% removal) vs log (time)*					
	Slope	0.095	0.1079	0.4353	0.5294	0.5622
	Correlation coefficient (*r*)	0.988	0.980	0.974	0.985	0.980
	Intercept	1.9754	1.695	0.924	0.7784	0.675

$$t/q_t = 1/k_2 q_e^2 + t/q_e \tag{9}$$

where, C_i and C_t = concentration of CH at time zero and time *t*, respectively (mg L^{-1}), q_e and q_t = amount of CH adsorbed at equilibrium time and time *t*, respectively(mg g^{-1}), $U(T) = [(C_i - C_t)/(C_i - C_e)]$, C_e = equilibrium CH concentration (mg L^{-1}), *k* = first order adsorption rate constant (min^{-1}).

The values of (i) $1/q_t$ (ii) $\log(C_i/C_t)$, (iii) $\log[1-U(T)]$ (iv) $\log(q_e-q_t)$ and (v) t/q_t were linearly correlated with time (*t*). The values of rate constant and correlation co-efficient (*r*-values) for all the above equations are given in Table 3. Fig. 3 represents pseudo second order rate equation. All the linear correlations were found to be statistically significant (*r*-values close to unity), indicating the applicability of these kinetic equations.

The experimental and calculated values of q_e for pseudo first order and pseudo second order equations are given in Table 4. The experimental q_e are found to be closer with calculated q_e values in the pseudo second order kinetic model. Similar report was given by Saravanan et al., on the biosorption of acid green using Rhodoturula Glutinis (Saravanan et al., 2012a,b). The R^2 value from Table 4 (0.999) also indicated the pseudo second order nature of adsorption of CH on these adsorbents.

In order to compare the validity of each model more efficiently a normalized standard deviation, Δ*q* (%) was calculated using the following equation:

$$\Delta q(\%) = 100 \times \left\{ \left(\Sigma \left[\left(q_t^{exp.} - q_t^{cal.} \right) / q_t^{exp.} \right]^2 \right) / (n-1) \right\}^{1/2} \tag{10}$$

where the superscripts, exp. and cal. are the experimental and calculated values of q_t *viz*., the amount adsorbed at different time t and n is the number of observations. The Δ*q* (%) values are also given in Table 4. Based on the low values of Δ*q* (%), it is concluded that the adsorption of chromotrope can best be described by the pseudo second order model.

3.3. Intra-particle diffusion model

The possibility of the intra-particle diffusion process was explored by using the Weber and Morris intra-particle diffusion model:

$$Q_t = k_p t^{1/2} + C \tag{11}$$

where q_t = amount of CH adsorbed at time, *t* (mg g^{-1}), *C* = intercept,k_p = intra-particle diffusion rate constant (mg g^{-1} min$^{-1/2}$).

The values of q_t were found to be linearly correlated with values of $t^{1/2}$. The k_p values were calculated by using correlation analysis. The *r*-values were found to be close to unity, indicating the application of this model. This revealed the presence of the intra-particle diffusion process. Similar report was given by Khan et al. on the adsorption of Rhodamine B on activated mango leaf carbon (Khan et al., 2011). The calculated values of k_p (mg g^{-1} min$^{-1/2}$) for adsorbents other than CAC are maximum for ASC (0.8222) and minimum for SNC (0.495). This result indicates that the intra-particle diffusion process is fast in ASC and slow in SNC. The values of intercept

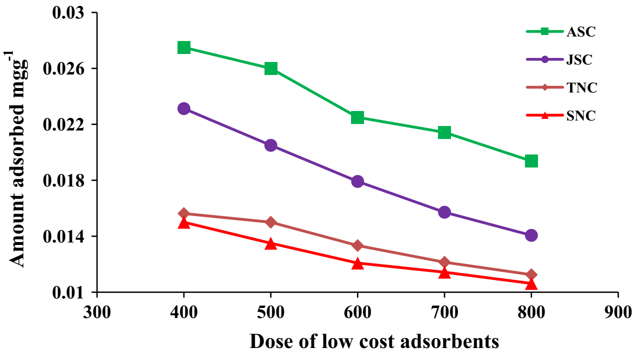

Figure 4 Effect of adsorbent dose variation on the adsorption of chromotrope on various carbons.

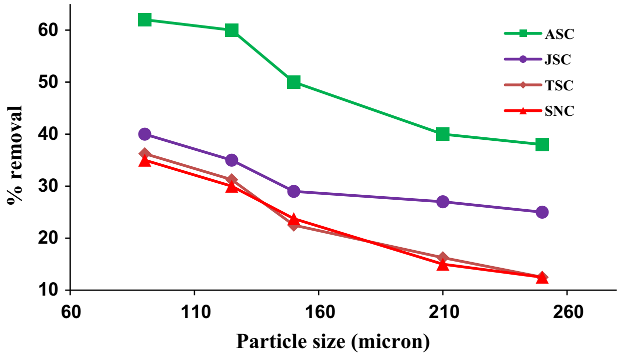

Figure 5 Effect of particle size variation on the adsorption of chromotrope on various carbons.

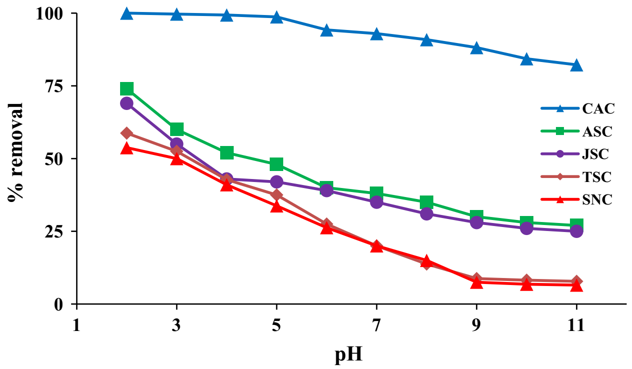

Figure 6 Effect of pH variation on the adsorption of chromotrope on various carbons.

(Table 5) give an idea about the boundary layer thickness i.e., the larger the value of C, the greater is the contribution of surface adsorption in the rate limiting step. The applicability of intra-particle diffusion is also confirmed by the lower values Δq (%).

$$\log(\%\text{removal}) = c + m\log(\text{time}) \quad (12)$$

where, c = intercept and m = slope. The correlation of the values of log (% removal) and log (time) also resulted in linear relationship which indicates that the process of intra-particle diffusion is taking place in these adsorption systems (Table 5).

3.4. Effect of adsorbent dosage

Fig. 4 shows the adsorption of CH as a function of adsorbent dosage. It is apparent that by increasing the adsorbent dose the amount of adsorbed dye increases but adsorption density, the amount adsorbed per unit mass, decreases. It is readily understood that the number of available adsorption sites increases by increasing the adsorbent dose and it, therefore, results in an increase of the amount of adsorbed dye. The decrease in adsorption density with an increase in the adsorbent dose is mainly because of unsaturation of adsorption sites through the adsorption process (Yu et al., 2003). Among the low cost carbons, the maximum amount of CH adsorbed is found to be 0.028 mg g^{-1} for ASC.

3.5. Effect of particle size

The effect of particle size was studied using CH on various carbons. As shown in Fig. 5, the percentage removal of CH increased with a decrease in the particle size. A decrease in the particle size would lead to an increase in surface area and the increase in the adsorption opportunity at the outer surface of the adsorbents. Besides adsorption at the outer surface of the adsorbent there is also the possibility of intraparticle diffusion from the outer surface into the pores of the material. The diffusional resistance to mass transfer is greater for large particles. Consequently the adsorption capacity of large particles

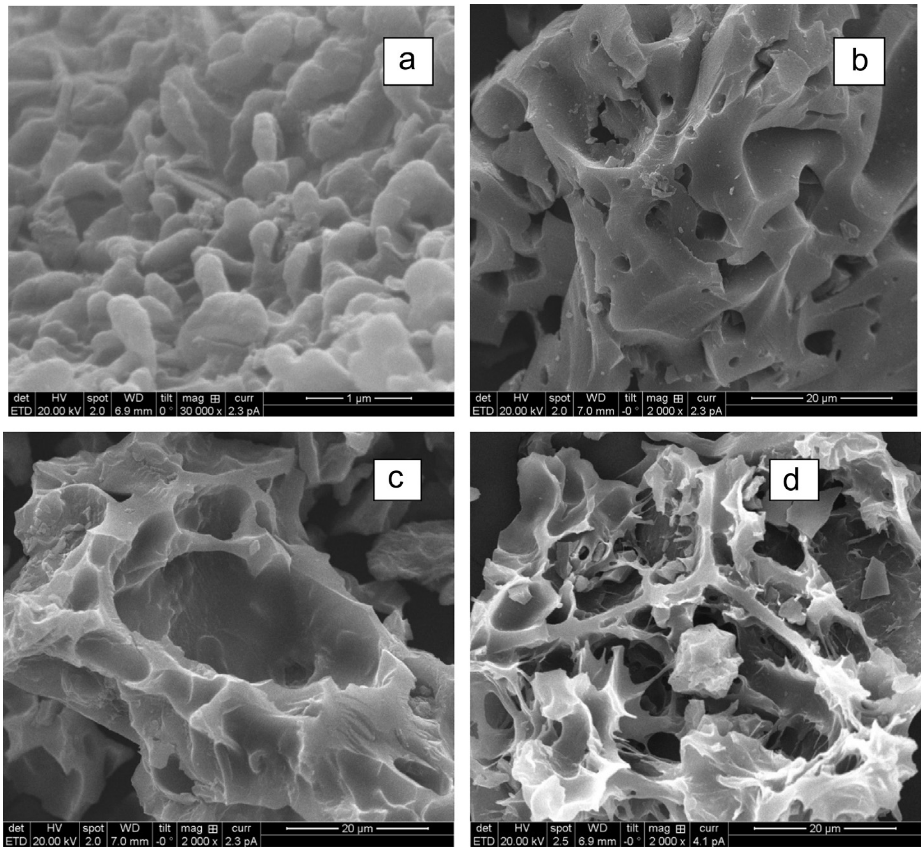

Figure 7 (a) SEM image of pure ASC. (b) SEM image of pure TSC. (c) SEM image of pure JSC. (d) SEM image of JSC loaded with chromotrope.

may be low (Ozacar and Sengil, 2002). ASC was found to have maximum percentage removal (62%) among all the low cost carbons.

3.6. *pH effect*

Effect of pH on the removal of CH was studied. The adsorption of this anionic dye onto the adsorbent surface is primarily influenced by the surface charge on the adsorbent, which in turn is influenced by the solution pH. The maximum removal for CH was observed at pH 2, when the surface is positively charged with excess protons in solution. Same kind of report was given by Kavitha et al. for the adsorption of acid brilliant blue (Kavitha and Namsivayam, 2008). Low pH value (1.0–3.0) leads to an increase in H^+ ion concentration in the system and the surface of the activated carbon acquires positive charge by absorbing H^+ ions. As the adsorbent surface is positively charged at low pH value, a significantly strong electrostatic attraction appears between the positively charged carbon surface and anionic dye molecule leading to maximum adsorption of chromotrope. Highest percentage removal of CH (74%) was observed for ASC at pH 2 among all the low cost carbons (Fig. 6).

3.7. *SEM analysis*

Scanning Electron Microscope (SEM) studies provide useful information regarding the textural/morphological characteristics of the activated carbons. SEM photographs (Fig. 7a and b) of the unloaded activated carbons show the rough and uneven surfaces of ASC and TSC. This surface property should be considered as a factor for dye binding. Fig. 7c is the picture of surface of unloaded JSC and Fig. 7d is the surface structure of CH loaded JSC. After dye adsorption, a significant change is observed in the structure of the adsorbent. The adsorbent appears to have a rough surface and pores containing new shiny particles after adsorption. Similar report was given for the adsorption of Rhodamine-B on parthenium biomass (Lata et al., 2008).

3.8. *FT-IR analysis*

Fourier transform infra-red spectra were recorded for the activated carbons with and without dye loading. The FTIR spectra of the low cost carbons are found to be almost similar with some peaks common, which are slightly shifted in the spectra of dye loaded activated carbons (Table 6). FT-IR spectra of pure ASC are given in Fig. 8a and FT-IR spectra of JSC and JSC loaded with dye are shown in Fig. 8b. The FT-IR spectra of pure ASC contain several peaks at 752.19, 1122.49, 1334.65, 1400.22, 1460.01, 1672.17, 1861.18, 3197.76 and 3313.48/cm, which confirm the presence of various functional groups on the surface of ASC. These surface functional groups are responsible for dye uptake capacity of adsorbents.

Table 6 FT-IR characteristic frequencies (ν in 1/cm) of ASC and JSC before and after adsorption of dyes.

Adsorbents	O–H stretching	–N = N– stretching	S = O stretching	C–S stretching
JSC	3197.76	1670.24	1120.56	754.12
CH on JSC	3313.48	1629.74	1124.42	750.26

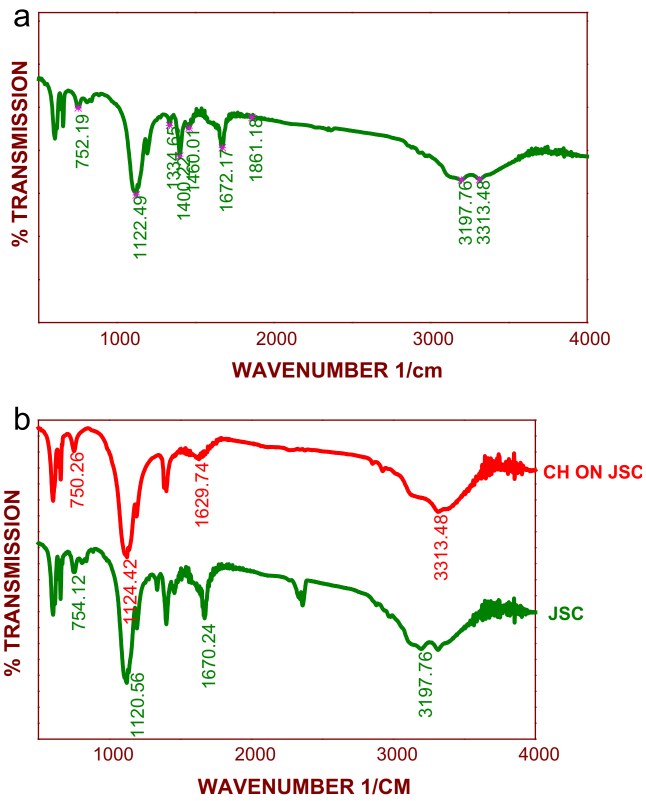

Figure 8 (a) FT-IR spectrum of unloaded ASC. (b) FT-IR spectrum of JSC and JSC loaded with dye chromotrope.

The peaks in the FT-IR spectra of unloaded JSC 3197.76, 1670.24, 1120.56 and 754.12/cm, may be due to O–H, –N=N–, S = O and C–S stretching, respectively. The peak at 1120.56/cm in unloaded JSC is shifted to 1124.42/cm on loaded with CH dye. The peak at 1670.24/cm in unloaded JSC is shifted to 1629.74/cm on loaded with CH dye. Similarly the peak around 750/cm in unloaded JSC is also slightly shifted on loaded with CH dye. Similar observation was made for the adsorption of heavy metals by a biopolymer (Saravanan et al., 2012a,b). Thus the FT-IR spectral analysis clearly indicates the adsorption of chromotrope dye onto the various activated carbons.

4. Conclusions

The results of the present investigation show that activated carbon prepared from low cost materials, aamla seed carbon (ASC), jambul seed carbon (JSC), tamarind seed carbon (TSC) and soapnut carbon (SNC) has suitable adsorption capacity with regard to the removal of chromotrope dye from its aqueous solution. Aamla seed carbon has better adsorption capacity than others. Thus it can be used as best alternative to CAC for the removal of chromotrope. The adsorption is highly dependent on adsorbent dose, particle size and pH. Adsorption obeys Langmuir and Redlich–Peterson equations better than Temkin, Freundlich and Dubinin–Radushkevich (D–R) equations. Adsorption kinetics fitted well onto pseudo second order rate equation. The applicability of the intra-particle model indicates the presence of the intra-particle diffusion process. Acidic pH is found to be more favourable for the chromotrope adsorption. FT-IR and SEM analysis clearly reveals the adsorption of chromotrope onto the activated carbons.

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