Amputate of Assorted Dyes from Polluted Aqueous Solution Using Novel Low Cost Adsorbents

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Abstract - **Application of activated carbons developed from cashew nut and Tamarind seed carbon from the agricultural waste product has been investigated for the removal of reactive magenta, reactive red, acid orange and crystal violet dyes from aqueous solution and waste water. Batch adsorption experiments are carried out for the removal of dyes from aqueous solution by using cashew nut and Tamarind seed carbon. The important parameters are studied like initial concentrations of dyes, contact time and dose of carbon for the removal of dyes. The data are fitted into Langmuir and Freundlich adsorption isotherm models and their constants are determined. For the kinetic study of the adsorption process under consideration, the following kinetic equations proposed by Natarajan and Khalaf, Lagergren, Bhattacharya and Venkobachar were employed. The intra- particle diffusion processes in often the rate limiting step in much obey first order nature of the adsorption process. The characterization of the physio chemically modified carbons are carried out by SEM, XRD analysis, UV-Visible and FT-IR analysis of before and after adsorption of dyes on the carbon surface area morphology studied.**
*Keywords***: Cashew nut carbon, Tamarind seed carbon, Adsorption isotherm, Kinetic study, Characterization**

I. INTRODUCTION

Water pollution due to discharge of colored effluents from textile dye manufacturing and textile dyeing mills are one of the major environmental concerns in the world today. Dyes important appearing colors to textile fibers, food stuffs, etc., however, the strong colors imported by the dyes pose aesthetic and ecological problems to the aquatic system. Because of their complexes molecular structures and large size of the dyes are considered non- oxidizable by conventional physical and biological process. Decolorization is one of the indispensable processes in waste water treatment. Among this physico chemical method like adsorption, electro chemical co-agulation and photo catalytic decolorization are more popular methods in nowadays. Among these methods adsorption is one of the methods which are gaining more and more attention because of its easy operation versatility. Waste water containg dyes containg dyes present a series environmental problem because it's high toxicity and possible accumulation in the environmental. Many of the dyes are synthetic and contain aromatic rings which make them carcinogenic and mutagenic therefore the removal of dyes from textile effluents is currently of great interest. Activated

carbon is most popular widely used in adsorbent is adsorption process(Arulanandam et., al 2015) because it has large surface area and it high adsorption capacity a number of low cost adsorbent have been used for the dye removal , adsorbent for including agricultural waste carbon likes viz., CNC and TSC.

Where used as the removal of dyes namely reactive magenta, reactive red, acid orange and crystal violet dyes. Aim of the present work is to explore the possibility of using this Carbonaceous materials as low cost adsorbents for these removal of dyes for waste water effective parameters such as initial dye conc. Adsorbent dosage, contact time, kinetics and various equations and isotherm model were used evaluate the adsorption of various activated carbon prepare from agricultural waste. The reactive dyes, which represent the largest class of dyes used in processing industries, are almost azo compounds, have molecules with one or several azo (N=N) bridges linking substituted aromatic structures. These dyes are designed to be chemically and photolytically stable; they exhibit a high resistance to microbial degradation and are highly persistent in natural environment (Suteu D., et al., 2005). Many of these dyes are also toxic and can cause dermatitis, skin irritation (Brookstein D.S, 2009) and also provoke cancer (de Lima R.O.A., et al., 2007) and mutation in humans (Carneiro P.A., et al., 2010) even carcinogenic and this poses a serious hazard to aquatic living organisms(ˊNeill C. O, et al., 1999; Vandevivere P.C., *et al.,* 1989). Due to this, it is important to remove these pollutants from the organic content of industrial effluents, it is necessary to eliminate dyes from wastewater before it is discharged (Machadoa F.M., *et al.*, 2011)

II. EXPERIMENTAL DETAILS

A. Dye solution preparation

These dyes viz., Reactive red (RR) [C.I name = Reactive Red 2, molecular weight = 615.33, λ_{max}=526 nm], Reactive Magenta (RM) [C.I. name = Reactive Magenta, molecular weight =1418.94, λ_{max}=544 nm], Acid Orange 8 (AO) [C.I name = Acid orange, Molecular weight =364.45, λ_{max} =483 nm] and Crystal Violet(CV) [C.I name = Crystal Violet, Molecular weight =407.99, λ_{max} =590 nm] were used as such without further purification. An accurately weighed

quantity of dyes was dissolved in double distilled water to prepare the stock solutions. The structure of dyes shown on the Figure-1a for Reactive red, 1b for Reactive Magenta 1c for Crystal Violet, and 1d for Acid Orange respectively. Nature and Physical and Chemical properties of dyes shown in Table 1.

TABLE 1 NATURE AND PHYSICAL PROPERTIES AND USES OF RPO AND RM DYES

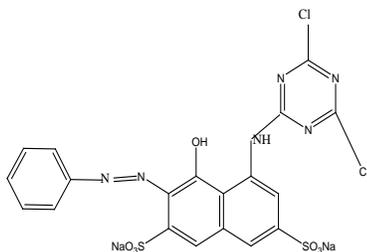| Nature of dye	Reactive Red	Reactive magenta	Acid Orange	Crystal violet
Generic name	Reactive brilliant Red X-3B	ReactiveMagenta MB	Acid Orange 2R	Methyl violet 10B
Color index No	12226-03-8)	42510	15575	-
Abbreviations	RR	RM	AO	CV
Commercial name	Reactive red 2	Reactive Magenta	Acid Orange 8	Crystal violet or gentian violet
Molecular formula	$C_{19}H_{10}Cl_2N_6Na_2O_7S_2$	$C_{27}H_{16}ClN_7Na_4O_{13}S_3$	$C_{17}H_{13}N_2NaO_4S$	$C_{25}H_{31}N_3OCl$
Purity	75%	87.86%	65%	82%
Chromophores	Single azo class	Single azo class	Single azo class	Single azo class
Molecular Weight	615.33424 [g/mol]	870.07	364.35	407.99
λmax-Value	526nm	544m	364.35nm	590 nm
Appearance	Rosy Red in color	Basic Violet -14	Orange powder	Dark green powder
Properties	Purplish red powder Water solubility	Dark purple powder, water solubility nature	For red orange is soluble in water, soluble in soluble fiber element, slightly soluble in ethanol and acetone, insoluble in other organic solvents	Powder nature, water solubility
Uses	Mainly used for cotton, viscose, wool, silk, polyamide fiber disseminated and roll dye, also can be used to knot dyeing. Storage stability is poorer.	dyeing of cotton silk, paper, leather and also in paint and printing inks	Used for wool, silk, cotton, vinegar, polyamide fiber and fiber dyeing. Also used for paper, leather dyeing, the heavy metal salt used in organic pigments.	a histological stain and in Gram's method of classifying bacteria antibacterial, antifungal, and anthelmintic properties and was formerly important as a topical antiseptic

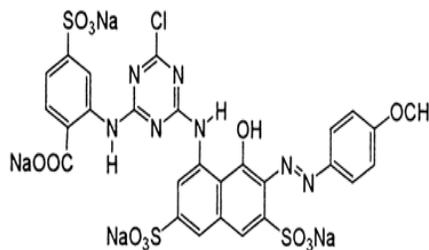Fig.1a Structure Of Reactive Red Dye
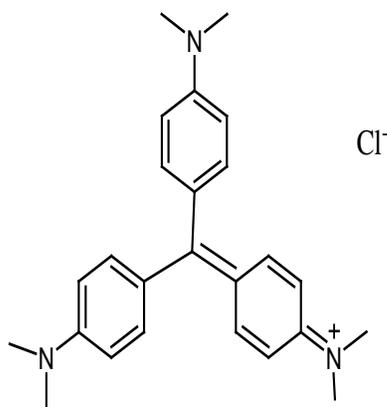

Fig.1b structure of Reactive Magenta
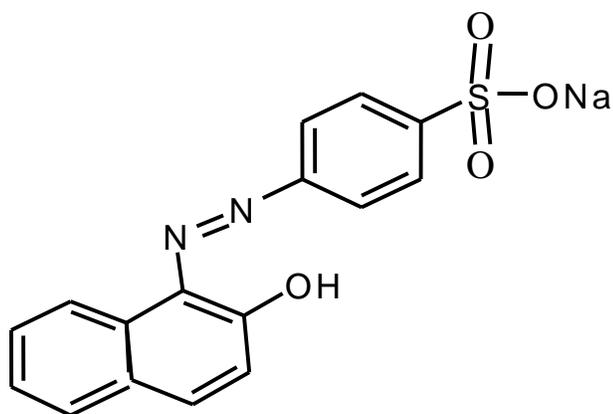
Fig.1c Structure of Crystal Violet

Fig.1d structure of Acid Orange

B. Adsorbent

Cashew nut shell and Tamarind seed carbons were used as adsorbents in this study. These carbons obtained from agricultural waste, which were washed with tap water and finally with double distilled water to remove the suspended impurities, like dust and soil and then dried in an air oven. About 500 g of powder Cashew nut shell carbon and Tamarind seed carbons was mixed with 100ml of con. Sulfuric acid separately and kept at room temperature for 24hrs. Then, it was dried in a hot air oven at 80˚C. The dried material was washed with distilled water to remove excess of acid. These activated Carbons were dried at 110˚C for 12 hours to remove moisture and kept in an air tightened bottle.

C. Adsorption Experiments

Batch mode adsorption studies were carried out by adding certain amount of dose of adsorbent and 50ml of dye solutions of certain concentrations and contact time of shaker at 200 rpm. The samples were withdrawn from the shaker at predetermined time intervals and solutions were separated from the adsorbent. To determine the residual dye concentration, the absorbance of the supernatant solution was measured before and after treatment with double beam spectrophotometer (VISISCAN 167 Spectrophotometer). The initial dye concentrations in the test solution and the contact time were changed to investigate their effect on the adsorption kinetics. The amount of adsorbed and adsorption efficiency of % removal was calculated as follows:

Amount adsorbed (q)   = $(C_o - C_e)$ /m   (1)
Efficiency (%) removal = $(C_o - C_e)$ /$C_o \times 100$ (2)

Where, C_o is the initial concentration (mg/L), C_e is the equilibrium concentration (mg/L), m is the mass of adsorbent (g), q is the amount adsorbed (mg/g).

III. RESULT AND DISCUSSION

A. Effect of concentration variation

In order to study the effect of initial concentration of RG and RR varying (40 - 75 ppm) and AO and CV varying (50-75 ppm) respectively and fixed amount of dose and contact time on the extent of the removal of dyes in terms of the percentage removal and amount adsorbed (mg/g) by using various carbons (CNC and TMC). The batch adsorption experiments where performed with the fixed dose of 20g/L and constant contact time (45 min.) and particle size 90 microns at 32± 1^0c. Initial concentration (C_o) of dyes (40 – 70 ppm for RR and RM and 50-75 ppm for AO and CV). Equilibrium concentration (C_e) of dyes can be calculated with help of calibration curve (known concentration of dyes without carbon and the absorbance should be noted). The amounts of dyes were found to increase exponentially with increasing initial concentration of dyes. The concentration of dyes increases, the percentage removal of adsorption decreases. This is due to lack of available active site on the surface carbons, which will enhance the formation of unilayer on the surface of carbon. Further formation of layer of dye molecule on the surface of carbon is highly hindered due to repulsive force between the adsorbate (dye molecule) and adsorbent (carbon) in the bulk of the solution. We have to optimize the initial concentration of dyes and fixed as 40 ppm for RR, RM and 50 ppm for AO and CV. Adsorption capacity of varies carbons for the removal of dyes are found to be order as follows, CAC >TMC >CNC. This is shown in the Table (2) and Figure-2a and 2b respectively.

TABLE 2 EFFECT OF INITIAL CONCENTRATIONS OF RR AND RM ON CNC AND AO AND CV ON TMC

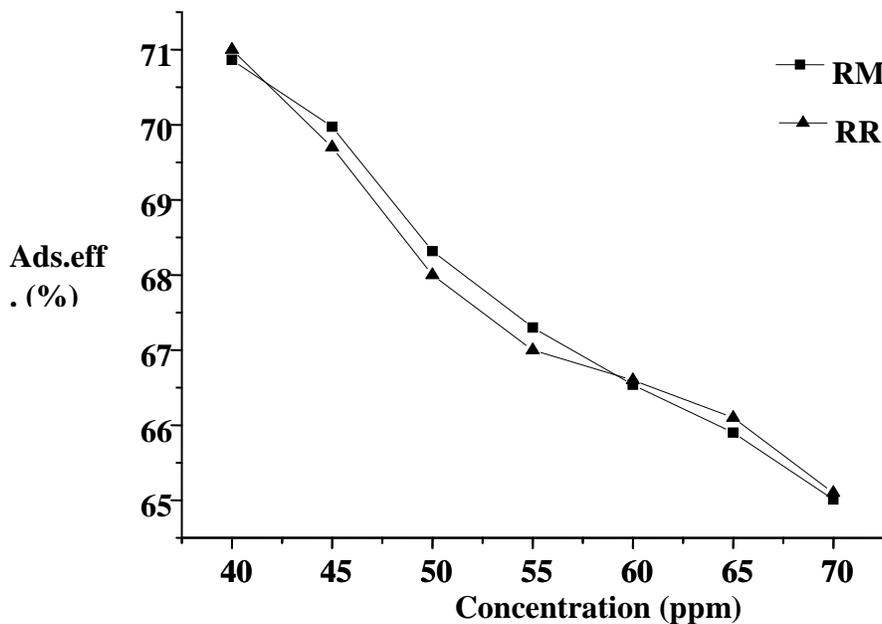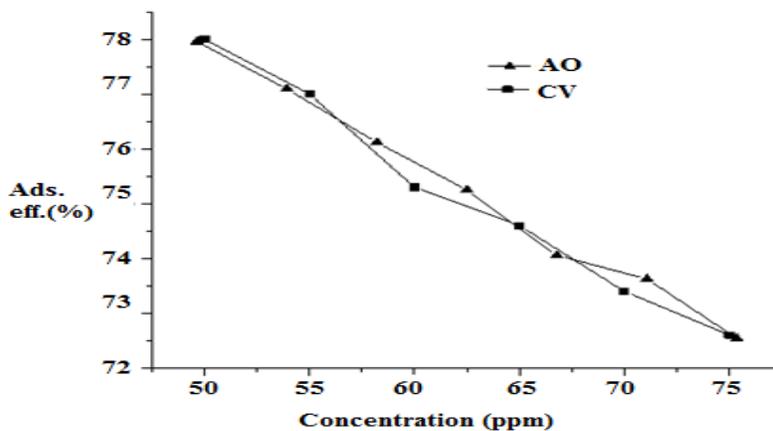| Conc. (ppm)	CNC				Conc. (ppm)	TMC			
	RR		RM			AO		CV	
	Ads. Eff.%	Amt. Ads.	Ads. Eff.%	Amt. Ads.		Ads. Eff.%	Amt. Ads.	Ads. Eff.%	Amt. Ads.
40	71	1.42	70	1.4	40	78	1.95	80	2.0
45	69.7	1.57	69.3	1.56	45	77	2.12	79.2	2.18
50`	68	1.75	68	1.7	50	75.3	2.26	78.3	2.35
55	67	1.85	67.2	1.85	55	74.6	2.42	77.5	2.52
60	66.6	1.99	66.6	2.0	60	73.4	2.57	76.4	2.68
65	66.1	2.15	66.1	2.15	65	72.6	2.72	76	2.85

Fig.2a Effect of initial Concentration for CNC

Fig.2b Effect of initial Concentration TMC

B. Adsorption isotherms

The applicability of adsorption isotherms has been important and significant for water and waste water treatment by adsorption technique. These isotherms are estimating the adsorption capacity of various carbons with respect to CAC as standard one. This shown in the Figures-3a and 3b for Freundlich isotherm, 3c and 3d for Langmuir isotherm respectively.

Freundlich isotherm

$$X/m = q_e = KC_e^{1/n} \qquad (3)$$

X -is the amount adsorbed in ppm $(X=C_0-C_e)$, m -is the mass of adsorbent (g/L) ,C_e -is the equilibrium concentration in ppm, K -is the Freundlich constant (to measure the adsorption capacity) 1/n – to measure the adsorption intensity.

The log x/m Vs log C_e graph should be linear in all cases. Values of 1/n for the dye substances indicate the adsorption mechanism with intra particle diffusion was the rate determined step and suggested by Weber and Morris, (W.J. Weber., and J.C. Morris et al., 1963)

$$q_t = K_p t^{1/2} + C \qquad (4)$$

q_t . is the amount adsorbed dye at a time t,C - is the intercept,K - is the intra particle diffusion rate constant.
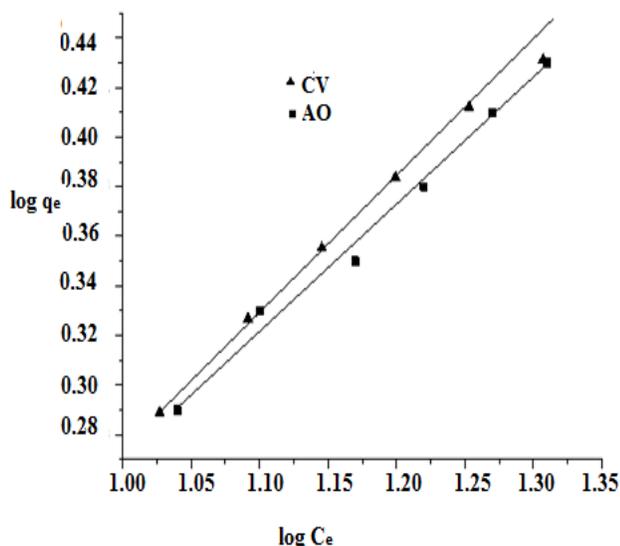

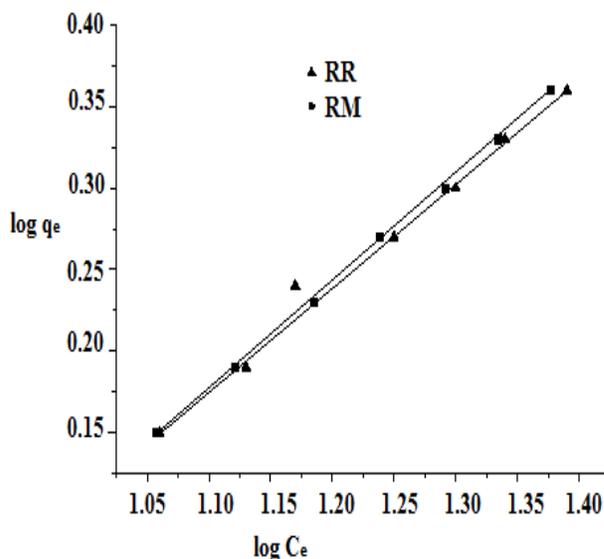Fig.3a Freundlich diagram for CV and AO for TMC

Fig.3b Freundlich diagram for RR and RM on TM

Langmuir isotherm

$$C_e/q_e = (1/ab) + C_e/a \qquad (5)$$

q_e is the amount adsorbed,C_e is the equilibrium concentration, a and b are Langmuir constant, a is the measure the adsorption capacity for the monolayer coverage, b is the energy of adsorption, We can draw a plot of C_e/q_e Vs C_e will be straight line with 1/ab values intercept and 1/a value is slope.

C. Effect of contact time

In order to study the kinetics and dynamics for adsorption of dyes using various carbons by the fixed amount of initial concentration (40 ppm for RR and RM) and dose (20g/L) by varying the contact time (30 to 60 min.), Particle size 90 microns at 32 ± 1^0C and its solution pH. These stop watch clock was started simultaneously during the starting of the experiment. The bottles were withdrawn from the shaker at the different time of (30 to 60 min.). Initially the percentage removal increases with increase in contact time to obtain maximum adsorption at 45 minutes after that to attain stagnated. When time goes on increasing upto optimum time and then the adsorption efficiency decreases due to desorption. This shown in the Table (3) and Figure.5a effect of contact time for RR and RM on CNC, 5b for CV and AO on TMC respectively.
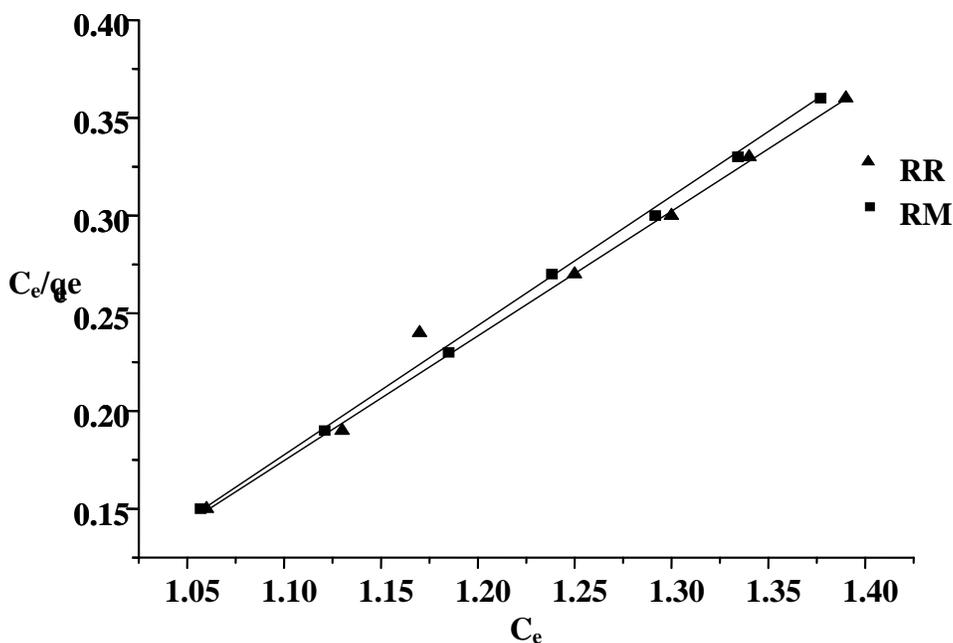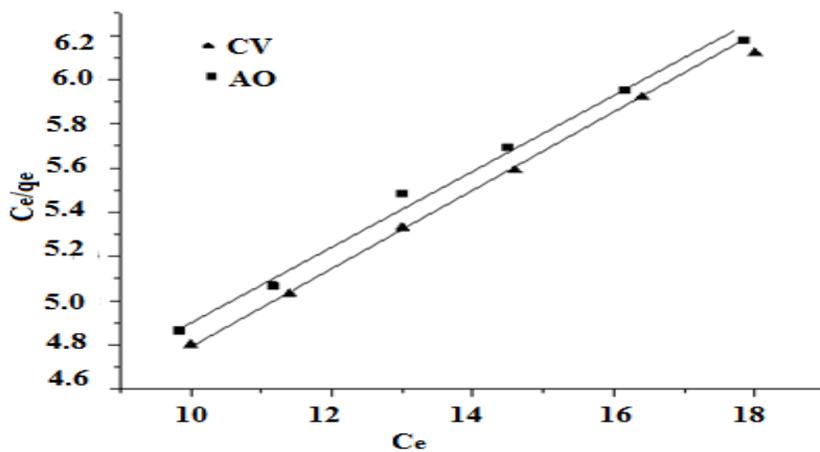
Fig.4a Langmuir isotherm for RR and RM on CNC

Fig.4b Langmuir isotherm for AO and CV on TMC

TABLE 3 EFFECT OF CONTACT TIME FOR CNC AND TMC

Time (Min.)	CNC				Time (Min.)	TMC			
	RR		RM			AO		CV	
	Ads. Eff. (%)	Amt. Ads.	Ads. Eff. (%)	Amt. Ads.		Ads. Eff. (%)	Amt. Ads.	Ads. Eff. (%)	Amt. Ads.
30	69.7	1.39	67	1.34	30	75.2	1.88	78	1.95
35	70.0	1.40	68.5	1.37	35	76.4	1.91	78.4	1.96
40	70.5	1.41	69	1.38	40	77.6	1.94	79.2	1.98
45	71.5	1.43	70	1.4	45	78.4	1.96	80.4	2.01
50	72	1.44	71	1.42	50	78.8	1.97	80.8	2.02
55	72.5	1.45	71.5	1.43	55	78.8	1.97	80.8	2.02
60	73	1.46	72	1.44	60	79.2	1.97	81.2	2.03

1.Kinetic isotherm models

The kinetic equations and intra particle model have been tested in our study. For the kinetic study of the adsorption process under consideration, the following kinetic equations proposed by Natarajan and Khalaf (Natarajan G.S.,et al., 1986)-as cited by Kannan and Vanangamudi (Kannan N.,et al., 1991), Lagergren equation (Lagergren S.,et al., 1984)-as cited by Panday, et al. (Panday K.K., et al., 1984) and Bhattacharya and Venkobachar equation (Bhattacharya A.K.,., et al., 1984) were employed.

Natarajan and Khalaf eqn. : $\log (C_i/C_t) = (k/2.303)\ t$ (6)
Lagergren eqn.   : $\log (q_e - q_t) = \log q_e - (k_{ad}/2.303)\ t$ (7)
Bhattacharya and
    Venkobachar eqn. : $\log [1-U\ (T)] = - (k_{ad}/2.303)\ t$ (8)

Where C_i and C_t are respectively the concentration of carboxylic acid (in ppm) at time zero and at time t; qe and qt are the amount of dye adsorbed per unit mass of the adsorbent (in mgg-1), respectively at equilibrium time and at time t; $U\ (T) = (C_i - C_t) / (C_o - C_e)$ and C_e is the equilibrium concentration of a dye (in ppm); k and k_{ad} are the adsorption rate constants (min^{-1}). The linear graphical plots between the values of $\log (C_i/C_t)$ and time, $\log (q_e - q_t)$ and time and $\log [1-U\ (T)]$ and time indicate the applicability of the above kinetic equations, and our system obeys the first order nature of the adsorption process. The Fig.6a for Lagergren plot for RR and RM on CNC and Fig.6b for CV and AO on TMC. Fig.6c Bhattacharya plot for RR, RM on CNC and 6d for CV, AO on TMC. Fig.6e and 6f for Natarajan Khalaf plot of CNC and TMC.

2.Intra-particle diffusion

The intra- particle diffusion process in often the rate limiting step in much adsorption process (McKay G., et al., 1980; Deo N. et al., 1993; Annadurai G et al., 1991; Annadurai G et al., 1998). The intra-particle diffusion model is given by the equation:

$$q_t = k_p\ t^{1/2} + c \qquad (9)$$

Where q_t is the amount of dye adsorbed (in mgg^{-1}) at time t; c and k_p are the intercept and intra-particle diffusion rate constant (unit: mgg^{-1} min$^{0.5}$), respectively. The graph of intra-particle diffusion shown in Fig.7a and 7b for Intra particle diffusion for RR, RM for CNC and CV, AO for TMC respectively.

C. Effect of adsorbent mass

The percentage removal and amount adsorbed dyes on the surface of carbon where obtained with different carbon dose of range (0.4g-1.6g) with fixed amount of initial concentration (40 ppm for RG and 40ppm for RR) and contact time (45 Min.) for CNC and initial concentration (50 ppm for AO and CV) for TMC. The percentage removal of dyes increase with increase in the amount of dose due to increasing available active site on the surface of carbon. From the study reveals that optimize the amount of dose of absorbents (carbons) 20g/L. These shown as Fig.8a for Effect of dose variation for RR and RM on CNC and Fig.8b for dose variation of AO and CV on TMC. These values are below the table-(4)

TABLE 4 EFFECT OF ADSORBENT DOSE FOR RR AND RM ON CNC & AO AND CV ON TMC

| Dose (g L^{-1}) | CNC | | | | Dose (g L^{-1}) | TMC | | | |
| | RR | | RM | | | AO | | CV | |
	Ads. Eff. (%)	Amt. Ads.	Ads. Eff. (%)	Amt. Ads.		Ads. Eff. (%)	Amt. Ads.	Ads. Eff. (%)	Amt. Ads.
0.4	68.1	1.36	68.5	1.37	0.4	74.8	1.87	77.6	1.94
0.6	69	1.38	69.5	1.39	0.6	76.8	1.92	78.8	1.97
0.8	70	1.4	69.5	1.39	0.8	77.2	1.93	79.6	1.99
1.0	71	1.42	70	1.4	1.0	78.4	1.	80.0	2.0
1.2	71	1.42	70.5	1.41	1.2	78.8	1.97	80.4	2.01
1.4	71.5	1.43	71.5	1.43	1.4	79.2	1.98	80.8	2.02
1.6	72	1.44	71.5	1.43	1.6	79.6	1.99	80.8	2.02

E. Spectrum analysis

1.FT-IR Spectra

FT-IR technique is an interesting application for studying the interaction between an adsorbate (dyes) and the active groups on the surface of adsorbent (P. Monash, et al., 2009). FT-IR spectra of before and after adsorption of CNC carbon and TMC carbon as shown in the Fig-9a and 9b respectively. A strong band at 1621 cm^{-1} for N=N azo dye stretching, 2276 cm^{-1} at Si-H stretch, 2341.58 cm^{-1} broad N-H stretch these additional peaks are observed due to the presence of dye on the surface of activated carbon (TMC). This shows that IR is the analytical technique to ensure the adsorption take place.

2.Electronic Spectra

The λ_{max} value for Reactive red 526 nm, Reactive Magenta λ_{max}=544 nm, Acid Orange λ_{max} =483 nm and Crystal Violet λ_{max} =590 nm was identified by the spectrum of UV-visible spectrum. These shown as Fig.10a, 10b, 10c, and 10d for UV-Visible spectrum of RR, RM, AO and CV respectively.

3.Scanning Electronic Microscopic

SEM is widely used for the study of morphological features and surface characteristics of the adsorbent materials (M.Arami, N.Y. et al., 2006) and then physical properties. It is useful for determining the particle shape, porosity and appropriate size distribution of the adsorbent (Ghosh,P.K.&Liji Philip,2005). In present study, SEM photographs of adsorbents for free carbon & after dye adsorbed carbon of CNC and TMC shown below in the Fig-11a, 11b, 11c and 11d from this analysis, it is clear that the appearance of surface is a microcrystalline structure.

4.XRD (X-Ray Diffraction method)

The Surface area is increases with affinity of the removal of dye due to presence of alkali and metal oxides on the surface of carbon. The XRD data for before and after adsorption of carbon CNC and TMC are shown below Fig-12a, 12b, 12c and 12d respectively.

IV. CONCLUSIONS

Activated carbons from agricultural by products/ wastes are used as adsorbents. Which are found to possess the essentially needed physico-chemical and textural properties of an adsorbent and hence they can be used as an adsorbent materials. The percentage removal of dyes is found to increase with the decrease in initial concentration of dyes increase with the increase in contact time and dosage of adsorbent. The process parameters were optimized. The relative adsorption capacities of the activated carbons has the maximum adsorption capacity, cost effective adsorbents which are alternatives to CAC in water and waste water treatment, especially for the removal of pollutants in general and dyes in particular. The adsorption data obeyed Freundlich and Langmuir adsorption isotherms indicating the formation of monolayer of dye molecules on the surface of the adsorbent. The adsorption data were modeled with the various kinetic equations and models such as Natarajan, Khalaf, Lagergren, Bhattacharya and Venkobachar equation and intra particle diffusion model. These models are found to be applicable. The result of the linear regression analysis of adsorption data revealed that the adsorption process is first order with intra particle diffusion as one of the rate determining step.

Abbreviations

CNC-Cashew Nut shell Carbon
TMC-Tamarind seed Carbon
RM-Reactive Magenta
RR-Reactive Red
AO-Acid Orange
CV-Crystal violet
XRD- X-Ray Diffraction
SEM-Scanning Electron Microscope
Ads. Eff. -Adsorption efficiency
Amt. Ads.-Amount Adsorbed
Conc.-Concentration

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