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Utilization of Waste Cigarette Buds for the Removal of Reactive Dye from Wastewater

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ABSTRACT
An excessiveamount of textile wastewater containing various dyes is being discharged into the various water receiving bodies with or without any treatment every day.Collected waste cigarette buds were washed with hydrochloric acid solution to be used as an adsorbent to remove reactive dye from textile wastewater. The adsorption studies were performed as a function of different concentration of dye solution, pH, and different adsorbent doses. Equilibrium data were analyzed using Langmuir, Freundlich, and Temkinisotherm models. Based on correlation coefficient (R^2) value, Langmuir model was found to be suitable. The kinetics of the adsorption process followed the pseudo-second-order kinetic model. FTIR and SEM analysis of the studied adsorbent before and after adsorption were also performed. Industrial feasibility of acid washed waste cigarette buds was performed and 98% dye removal was observed at optimum condition of pH 5.0, adsorbent dose of 20 g/L and the initial dye concentration of 100 ppm.

KEYWORDS: Adsorption, Kinetics, Isotherm, Reactive Dye, Cigarette buds, Waste management.

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I. INTRODUCTION

Ecological balance is happening due toincreased environmental pollution caused by rapid industrialization in the world. Textile industries more specifically dying industries are one of many to be held responsible for this environmental pollution [1]. Water pollutions, as well as air pollutions, are being caused by these industriesby discharging a large amount of wastewater into various receiving bodies and by ejecting different gases into atmosphere respectively [2]. In fact, major pollution due to textile industries is discharging colored effluents containing various types of dyes. Because of complex structure, most of thesedyes are stable towards photo-degradation and biodegradation [3]which are harmful to the environmental beings. Many of the organic and inorganic dyes are hazardous and may affect aquatic life and the food chain as well. Furthermore, some dyes or their metabolites are toxic, mutagenic or carcinogenic in nature [4]. There is no argument that colored effluents discharged by textile industry are a dangerous source of environmental contamination not only from the aesthetic point of view but also due to its toxicity to aquatic life [5]. So it is crucial to remove dye from colored effluent for safe discharge in water receiving bodies and prevent water pollution due to textile effluent.

The conventional processes for the dye removal from wastewater include biological treatment, coagulation, flocculation, oxidation, microfiltration and hyper-filtration that require high capital investment and running cost [6]. On the other hand, adsorption, a physiochemical treatment process, has gained prominence as a means of producing high-quality effluents that are low in metal ion concentrations and dyes as well. Adsorption using commercial activated carbon for the removal of dye has found widespread in the treatment of industrial wastewater. However, the high cost of commercial activated carbon and its load on non-renewable resources are not affordable for many developing countries. Therefore, there is a need to produce activated carbon from cheaper and readily available materials from renewable sources [7].Preparation of adsorbents from agriculture waste and by-product has been widely conducted. A number of low-cost adsorbents, such as activated carbon prepared from various wastes [8; 9; 10] baggas fly ash [5], Rice Husk [11; 12], coal fly ash [13], Tendue leaves [14], Granulated carbon,powdered carbon have been studied for adsorption of dye.

In the present study waste cigarette buds are used as adsorbents for removal of reactive dye from textile wastewater. Cellulose acetate fiber is the main constituent of the cigarette filter [15]. It is one type of plastic that degrades very slowly in the environment. It has been estimated that under ideal conditions the cellulose acetate fibertakes up to 18 months to degrade [16]. These cellulose acetate fibers, each having the diameter of about 20, are treated with titanium dioxide and over 15,000 of them are packed tightly together, using glycerol triacetate as a binding agent, to create a single filter [17]. An estimated 5.6 trillion filtered cigarettes were consumed worldwide in 2002 and nine trillion are projected for 2025 [18]. With global cigarette consumption on the rise, the global environmental burden of cigarette litter could be significantlyintensified in the coming years unless there is a change in the manner in which global populations dispose of these cigarette buds. Due to the persistent nature and volume of cigarette buds discharged into the environment, it is high time to find out an alternative use of this discharged waste.

II. MATERIALS & METHODS

2.1 Adsorbent Preparation

Waste cigarette buds were collected from various areas of Dhaka Metropolitan City. The collected cigarette buds were first washed thoroughly five times with deionized water. Then they were squeezed by hand to get rid of excess water. After that, they were dried in an oven at a temperature of 105^{o}C for 2 hours to remove the bound moisture. Later they were washedby a 5% hydrochloric acid solution until the washing becomes clear. After that, they were again squeezed and then dried in an oven at 105^{o}C for another 2 hours.

2.2 Preparation of Standard Solution

LiyuansolRed 3BE-150% a reactive dye was collected from a local dyeing factory. 1000 ppm stock solution was prepared by dissolving 1.0 g of dye in 1L deionized water. The desired amount of solutions were prepared from this stock solution and diluted using deionized water. pHwas adjusted by adding 0.1 M sodium hydroxide solution and 0.1 M hydrochloric acid solution.

2.3 Batch Adsorption Study

Batch process was employed for adsorption studies. A pre-weighted sample of the adsorbent and a measured volume of dye solution were taken in a 250mL Erlenmeyer flask. The mixture was agitated at 180 rpm for 360 min to ensure equilibrium. The absorbance of the dyesolution was measured in a double beam spectrophotometer at 542 nm wavelength by Shimadzu UV- 1800 UV– spectrophotometer. All the experiments were carried out at least three times at ambient temperature (25^{o}C). The amount of adsorption at equilibrium time, q_e(mg/g) was calculated using the equation:

$$q_e = (C_o - C_e)V/W$$

Where, C_0 for initial dye concentration (mg/L); C_e, for the dye concentration at equilibrium (mg/L); V represents the volume of the solution, (L); W for mass of dry adsorbent used, (g).

III. RESULTS & DISCUSSION

3.1 pH Effect

The pH of the dye solution has played a great role in the dyes adsorption by waste cigarette buds. At pH 5.0 the maximum adsorption was attained by acid washed waste cigarette buds (Fig. 1). As the pH was increased the removal efficiency decreased. This is due to OH^- ions contending with the anionic charges of the dye solution for adsorption on active sites. Same can be explained for acidic medium, i.e. H^+ ions contending with the cationic charges of the dye solution.

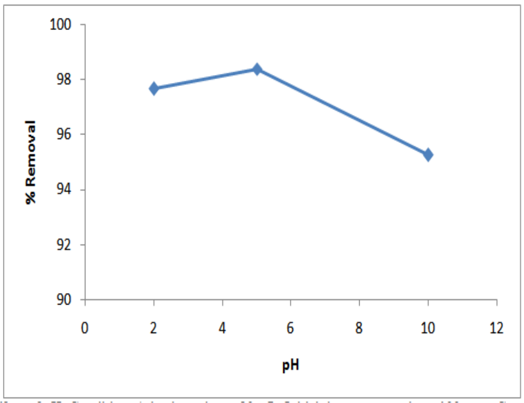


Fig. 1. Effect of pH. Condition: Adsorbent dose= 20 g/L, Initial dye concentration= 100 ppm, Contact time= 360 minutes, Agitation rate= 180 rpm, Temperature= 25^{o}C.

3.2 Adsorbent Dose Effect

The effect of adsorbent dosage was explored by varying the adsorbent dosages (20, 40, 60 and 80 g/L) at a fixed initial dye concentration of 100 ppm. The percent removal is presented in Fig. 2. When the adsorbent dosage was increased from 20 to 40 g/L, the percent removal remained fairly the same which was around 98.4%. Above 40 g/L, the removal efficiency decreased. It is a recognized fact that the percent removal increases rapidly with increase in the adsorbent dose because of the higher availability of the exchangeable sites or surface area. However, very slow increase in removal beyond an optimum dose may be observed becauseof the fulfillment of equilibrium between adsorbate and adsorbent at the existing operating conditions [19]. Screening effect of a dense outer layer of cells caused by higher adsorbent doses block the binding sites andresults in lower dye removal per unit adsorbent [20]. The optimum dose for the removal of reactive dye was chosen to be 20 g/L with removal efficiency of 98.39%.

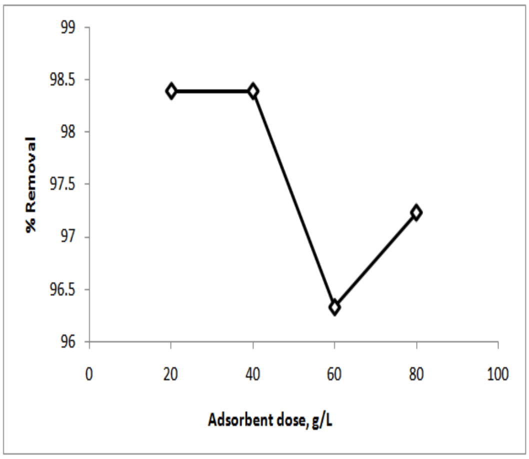


Fig. 2. Effect of adsorbent dose. Condition: pH= 5.0, Initial dye concentration= 100 ppm, Contact time= 360 minutes, Agitation rate= 180 rpm, Temperature= 25^{o}C.

3.3 Initial Dye Concentration Effect

Effect of initial dye concentration on its removal was carried out at an optimized adsorbent dose, contact time and pH by varying the dye concentration from 100-400 ppm (Fig. 3). This experiment was carried outfor the prediction of the driving force needed to overcome the mass transfer restriction of dye molecules between aqueous and solid surface [21;22].

Fig. 3 shows that the removal of reactive dyedecreases with increase in concentration from 100- 300 ppm. Because of the escalation in number of dye molecules competing for available binding sites and because ofscarcity of free binding sites[23] or because of the development of a barrier by the adsorbed dye molecules surrounding surface of the adsorbents, the dye removalpercentage decreases for the case of initial dye concentration of 200 ppm & 300 ppm. The increase in percent removal for the case of 400 ppm is due to the creation of newer binding sites by already adsorbed dye molecules or increased rate of intraparticle diffusion of dye molecules into the pores of the adsorbent [20].

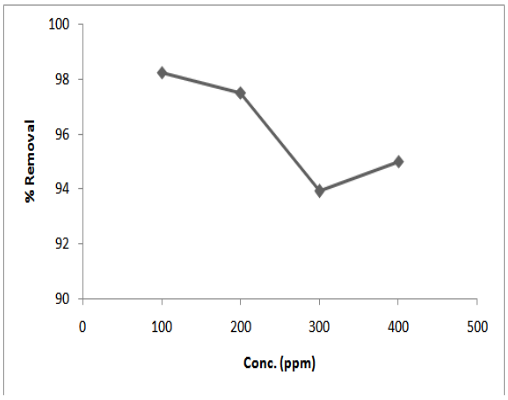


Fig. 3. Effect of initial dye concentration. Condition: pH= 5.0, Adsorbent dose= 20 g/L, Agitation rate= 180 rpm, Contact time= 360 mins, Temperature= 25^{o}C.

3.4 Adsorption Isotherm

Certain constant values whichnot only states the surface properties and affinity of an adsorbent but also utilized to compare the adsorptive capacities of that adsorbent for different pollutants can be termed as adsorption isotherm [24]. In order to predict the mechanism of dye adsorption on acid washed waste cigarette buds,the experimental data were applied to the Langmuir, Freundlich and Temkin isotherm equation. Using a linear form of the isotherm equations,the constant parameters of the isotherm equations for this adsorption process were calculated. The constant parameters and correlation coefficient (R^2) are presented in Table 1.

Langmuir Isotherm	
Q_o (mg/g)	21.008
K (L/g).	0.175
R^2	0.989
Freundlich Isotherm	
K_f (L/g)	3.976
n	2.045
R^2	0.95
Temkin Isotherm	
A (L/g)	1.502
b	502.693
R^2	0.908

Table 1. Isotherm constants for adsorption of reactive dye on acid washed waste cigarette buds.

3.4.1 Langmuir isotherm model:

The Langmuir isotherm adopts that the adsorption of molecules takes places as homogeneous sites, all sites are equivalent and there are no collaborations between adsorbate molecule and neighboring sites. The Langmuir isotherm equation in linear form is as follows,

$$1/q_e = (1/Q_oK)\,(1/C_e) + (1/Q_o)\ldots\ldots\ldots\ldots\ldots\ldots\ldots(1)$$

where, Ce is the equilibrium concentration (ppm), qe is the amount absorbed at equilibrium (mg/g), Q_0 and K is the Langmuir constants related to adsorption capacity and energy of adsorption.

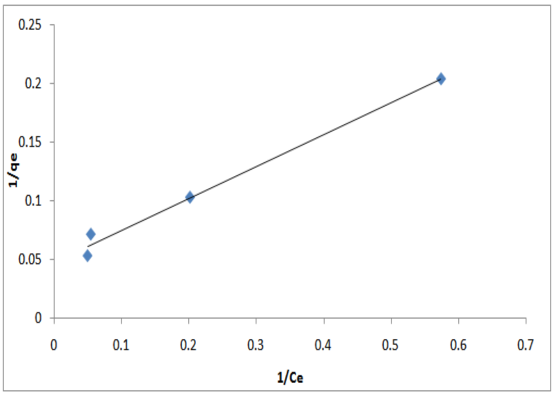


Fig. 4. Langmuir Isotherm.

Whether an adsorption system is “favorable” or “unfavorable”, can be predicted by a dimensionless constant, separation factor, R_L. The separation factor, R_L is defined by Eq. (2);

$R_L = 1/ (1 + KC_o)$……………………………….(2)

HereC$_o$ is the initial dye concentration (ppm) and K is the Langmuir adsorption equilibrium constant (L/g). The isotherm is considered unfavorable when $R_L > 1$, the isotherm is linear when $R_L = 1$, the isotherm is favorable when $0 < R_L < 1$ and the isotherm is irreversible when $R_L = 0$ [25]. $R_L$values were found to be 0.983, 0.966, 0.95 and 0.935 while initial dye concentrations were 100, 200, 300 and 400 ppm respectively, and therefore, favorable adsorption of dye molecules on the studied adsorbent can be stated.

3.4.2 Freundlich isotherm model:

The Freundlich isotherm adopts that the adsorption of molecules on the solid surface is proportional to its concentration in the aqueous medium at low pressure. Its linear form can be written as,

$\ln q_e = \ln K_f + (1/n) \ln C_e$ (3)

Here, q_e is the amount absorbed at equilibrium (mg/g), C_e is the equilibrium concentration (ppm),K_f and nbeing the Freundlich model constants where the value of n will be greater than 1.

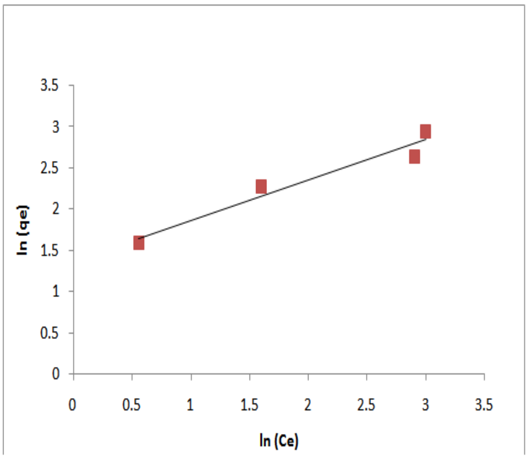


Fig. 5. Freundlich Isotherm.

3.4.3 Temkin isotherm:

The Temkin isotherm adopts that the fall in the heat of sorption is not logarithmic as referred in the Freundlich equation, rather, it is linear. The Temkin isotherm model is shown in Eq. (4):

$q_e = (RT/b) \ln (AC_e)$………………………….(4)

The linear form is shown in Eq. (5);

$q_e = B \ln A + B \ln C_e$………………………….(5)

Where B = (RT/b).q_e(mg/g) and C_e(ppm) is the amount absorbed at equilibrium and the equilibrium concentration, respectively. Also, T the absolute temperature in Kelvin (K) and R is the universal gas constant, 8.314 $\mathrm{Jmol^{-1}K^{-1}}$. The constant b is related to the heat of adsorption [26;27].

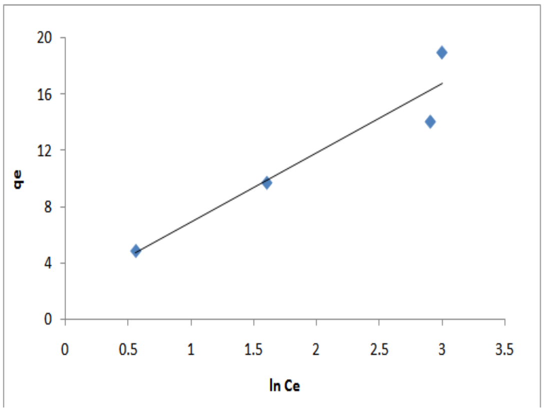


Fig. 6. Temkin Isotherm.

From Table 1 and Fig. 4 - 6, it can be said that the adsorption of reactive dye onto acid washed waste cigarette buds best fits into the Langmuir and Freundlich isotherm model. The experimental data also fits quite well into the Temkin isotherm model (all $R^2 > 0.9$).

As seen from Fig. 4, the Langmuir isotherm for adsorption of dye onto waste buds was found to be linear and the correlation coefficient was above 0.95 as tabulated in Table 1. From Fig. 5, it was seen that the Freundlich isotherm also fits well for dye adsorption by waste buds. The correlation coefficient was also satisfactory as shown in Table 1. The value of n was found to be greater than unity as shown in Table 1, representingfavorable adsorption of reactive dye by acid washed waste cigarette buds. It was seen from Fig. 6 that the Temkin isotherm for dye adsorption by waste buds was also found to be linear and the correlation coefficient was just above 0.9, as seen in Table 1. Since the Temkin isotherm constant (B) is related to the heat of adsorption, the adsorption of reactive dye onto acid washed cigarette buds could think of as a monolayer endothermic adsorption [20,21].

3.5 Adsorption Kinetics

Using pseudo first-order and pseudo-second-order kinetic models, the rate constants were calculated. The pseudo-first-order equation is as follows,

$\log (q_e - q_t) = \log q_e - (k_1/2.303)\, t$………………(6)

Where q_t(mg/g) is the amount of adsorbed dye molecules on the adsorbent at time t and k_1 (min^{-1}) is the rate constant of the first-order adsorption. Applicability of this kinetic model can be achieved if a straight line fromlog(q_e- q_t) versus tis obtained. q_e and k_1 can be determined from the intercept and slope of the plot respectively [28].

The pseudo-second-order kinetic model can be expressed as:

$t/q_t = (1/k_2 q_e^2) + (1/q_e)\, t$…………………………(7)

Where k_2 (g mg^{-1} min^{-1}) is the rate constant of second order adsorption. The pseudo-second-order kinetics will be applicable if the plot t/q_tversus t gives a straight line. Thenq_e and k_2 can be determined from the slope and intercept of the plot respectively.

Again,

$h = k_2 q_e^2$…………………………………………(8)

Where, h is the initial sorption rate (mg g^{-1} min^{-1})[29].

The pseudo first-order rate constants k_1, pseudo-second-order rate constants k_2, initial sorption rate h, calculated equilibrium sorption capacity $q_{e,theo}$(theoretical), experimental equilibrium sorption capacity $q_{e,\ exp}$(experimental) and regression coefficients R^2at various initial dye concentrations and at a fixed temperature (25°C) are given in Table 2.

C_o (ppm)	$q_{e,exp}$(mg /g)	Pseudo first- order kinetics			Pseudo second- order kinetics			
		k_1(min-1)	$q_{e,\ theo}$	R^2	k_2(g mg^{-1} min^{-1})	$q_{e,\ theo}$	h (mg g^{-1} min^{-1})	R^2
100	3.694	0.0005	1.047	0.007	0.0522	4.131	0.8905	0.949
200	6.996	0.0032	2.335	0.383	0.0168	7.564	0.9593	0.988
300	11.996	0.0117	8.236	0.784	0.0011	14.430	0.2331	0.944
400	19.025	0.0078	6.758	0.362	0.0014	20.284	0.5751	0.955

Table 2.A comparison of the pseudo first- order and pseudo-second-order rate constants and theoretical and experimental q_e values obtained at different initial dye concentrations and at fixed temperature 25°C.

A plot q_e versus initial dye concentration is shown in Fig. 7 for thecomparison of $q_{e,\ exp}$and $q_{e,\ theo}$results at various initial dye concentration.

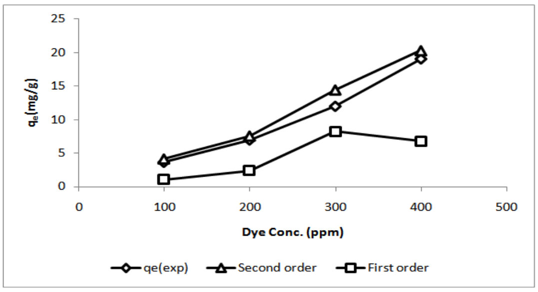


Fig. 7. Comparison between the experimental ($q_{e,\ exp}$) and calculated ($q_{e,\ theo}$) data for adsorption of reactive dye by acid washed waste cigarette buds at 25°C.

From Table 2 it can be seen that the pseudo-first-order kinetic model returns a bad correlation as regression coefficients were much below. On the other hand, all the regression coefficients for the pseudo-second-order kinetic model were found to be greater than 0.94 and thus indicates a good correlation. The q_e values determined from the first order kinetic model gave different values compared to that of experimental values as observed from Table 2 and Fig. 7 as well. However, for the case of the pseudo-second-order kinetic model the calculated $q_{e,\ theo}$values were quite close to $q_{e,\ exp}$values (± 6% to ± 20%) at various initial dye concentrations. Comparing the values of regression coefficients R^2, it was seen that the values for the pseudo-first-order model were very much lower than those of pseudo-second-order model for all the adsorption process and therefore it can be said that the pseudo second-order- model fits better than the pseudo-first-order model.

3.6 Fourier Transform Infrared Spectroscopy (FTIR) Analysis

The functional groups before and after adsorption of Liyuansol Red 3BE-150%, a reactive dye ontoacid washed waste cigarette buds and the corresponding infrared adsorption bands are shown in Table 3. Using Shimadzu IR Prestige- 21 FTIR spectrophotometerthe infrared spectra were obtained. The FTIR spectra before and after adsorption are shown in Fig. 8(a) and 8(b) respectively. It was clear that a strong intensity band should be present at 1616 cm-1indicating symmetric stretching of the carbonyl group (C=O) of the acetyl group (-COCH3) of cellulose acetate [30]. A reduction in the intensity of the band at 3475 cm-1 represents the hydroxyl group (-OH).

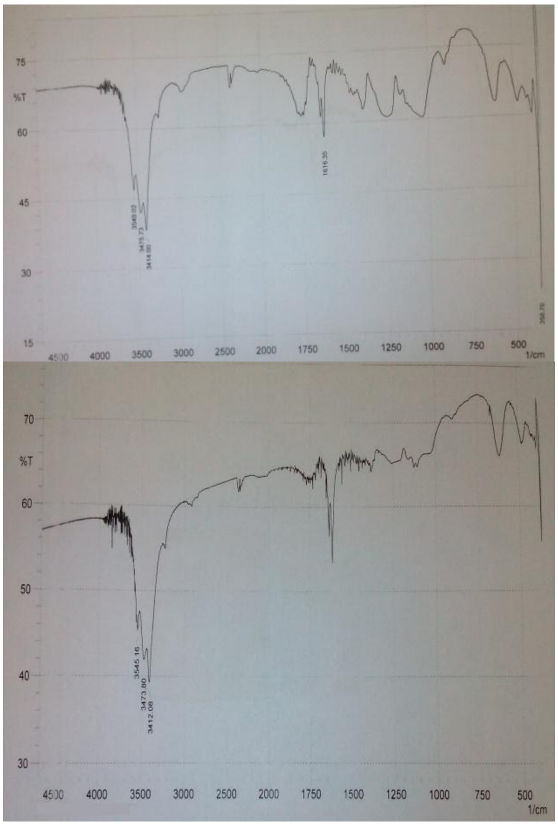


Fig. 8. (a) The FTIR spectra of acid washed waste cigarette buds before adsorption. (b) The FTIR spectra after adsorption. Condition: pH= 5.0, Initial dye concentration= 100 ppm, Contact time= 360 minutes, Agitation rate= 180 rpm, Temperature= 25°C.

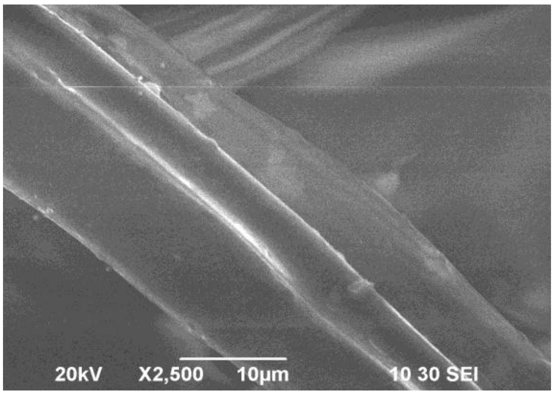
The dye molecules were mainly adsorbed by hydroxyl groups (-OH)as most of the peak shifts have occurred at that band [25].

IR peak	Adsorption Bands (cm^{-1})			Comments
	Before adsorption	After adsorption	Difference	
1	3549	3545	-4	Bonded –OH stretching
2	3475	3473	-2	Bonded –OH stretching
3	3414	3412	-2	Bonded –OH stretching

Table 3. FTIR spectral characteristics of acid washed waste cigarette buds before and after adsorption of reactive dye.

3.7 Scanning Electron Microscope (SEM) Analysis

Scanning Electron Microscope (SEM) study is generally used to investigate the morphology and surface characteristics of an adsorbent [31].SEM pictures of acid washed waste cigarette buds before and after adsorption of reactive dye molecules (Liyuansol Red 3BE-150%) are shown in Fig. 9(a) and 9(b) respectively. The surface of the studied adsorbent appears to be smooth before adsorption of dye molecules as seen in Fig. 9(a) whereas in Fig. 9(b) the dye molecules are being deposited on the surface of the adsorbent after adsorption.



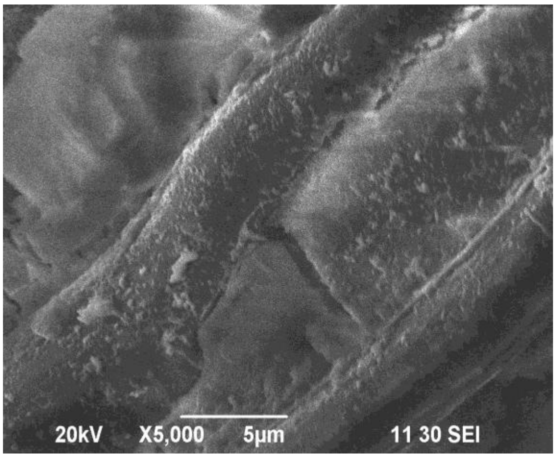


Fig. 9. (a) SEM of acid washed waste cigarette buds before adsorption. (b) SEM after adsorption.

3.8 Industrial Feasibility of Adsorbent

The industrial feasibility of acid washed waste cigarette buds was studied for the removal of dye from textile effluent at optimized conditions (pH= 5.0, Adsorbent dose= 20 g/L, Initial dye Concentration= 100 ppm, Agitation rate= 180 rpm, Contact time= 360 mins, Temperature= 25^oC)obtained from batch experiments. The removal efficiency of the adsorbent was outstanding and percent removal was found up to98.0 %.

IV. CONCLUSION

The adsorption of Liyuansol Red 3BE-150%, a reactive dye, by acid washed waste cigarette buds was dependent on pH, adsorbent dose, and initial dye concentration. The maximum dye removal was obtained at the optimal condition of pH 5.0, adsorbent dose of 20 g/L and initial concentration of 100 ppm. The waste buds were found suitable at the optimal conditions for the removal of reactive dyes from its industrial effluents as per percent removal is the concern (98.0%). The adsorption mechanism of reactive dye onto acid washed waste cigarette buds were defined by using Langmuir, Freundlich and Temkin isotherm and it was found to be an endothermic monolayer adsorption.SEM analysis also supports this statement as the dye molecules were spotted on the surface of the fibers. The pseudo first- order and pseudo-second-order kinetic model were used to analyze the data obtained for the adsorption of studied reactive dye onto acid washed waste cigarette buds. The pseudo-second-order equation was found to have the best correlation for the adsorption data.But the question still remains at large, whether the utilization of this waste cigarette buds as adsorbent exert any eco-toxic effect on marine and aquatic environment. Further research is suggested to explore the actual risks that cigarette waste has on freshwater and marine environments.

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