Efficient Removal of Methyl Orange and Rhodamine-B Dyes with Low Cost Banana Peel Activated Carbon

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Acid activated carbon obtained from cheap, non-toxic and locally available banana peel was used as a low cost and efficient adsorbent for the removal of dyes methyl orange and rhodamine-B from the aqueous solution. Changes in the resulting material before and after activation and after treatment were studied by different techniques, such as SEM-EDX, XRD, FTIR measurements. Effects of duration of treatment, amount of banana peel activated carbon, pH, and initial methyl orange and rhodamine-B concentration, on the removal of dye were studied to get optimum conditions for maximum dye removal. Removal efficiency of the activated ash remains almost constant in a wide range of pH from 2.5 to 5.6. In 75 min at room temperature removal of 98.5 % methyl orange (anionic) and 99.0 % rhodamine-B (cationic) dyes with 0.1 g and 0.125 g, respectively was obtained from the contaminated water having 10 ppm dye concentration.

Keywords: Activated carbon, Banana peel, Adsorbent, Dye, Methyl orange, Rhodamine-B.

INTRODUCTION

With the recent industrialization of especially the developing countries, organic dyes have become the major sources of pollution of surface water. Organic dyes are extensively used in many industries like leather, textile, paper, printing, *etc*. [1]. About 15 % of the dye is wasted and released as the untreated effluent. Removal of organic dyes is rather difficult due to aromatic structure of pollutants and their non-degradable synthetic origins. Azo dyes, the largest class of synthetic dyes characterized by the presence of nitrogen double bonds (-N=N-), are well known mutants, carcinogens for human beings and toxic to aquatic organisms [2] and their removal from effluent is necessary before their discharge into the water streams. Various techniques like chemical coagulation/flocculation [3,4], ozonation [5], oxidation processes [6], ultrafiltration [7]. Most of these methods have certain limitations in their applications, for example very harsh reaction conditions, high cost [8], secondary pollution and intensive energy requirements. Adsorption has a promising prospect and a wide range of applications due to its high removal efficiency [9], low cost, mild operating conditions, no secondary pollution and good performance over other conventional treatment processes in the removal of dyes from wastewaters. Although adsorption processes using activated carbons have been found to be effective in the treatment of these waste waters [10] due to its increased surface area, micropore structure, high adsorption kinetics with low cost and greater potential for methyl orange removal from waste water, the preparation and regeneration of activated carbons are relatively costly, thus limiting their application. Therefore, interest in the production of low-cost natural waste materials for dye removal has increased considerably [11-13].

Agricultural waste materials have little or no economic value and often pose a disposal problem. Extraction of sorbents from agricultural wastes is of great importance as it is cost effective. Many agricultural byproducts had been locally available for synthesis of activated carbon and used for dye sorption from wastewaters. The activated carbon are mutable adsorbent and have specific adsorption capacity property which depends on its pore size and surface area, which include wood apple shell [14], green coconut fibers [15], peanut hull [16], rice husk [17], wood sawdust [18], peanut husk [19], pine sawdust [20], banana peel and green coconut mesocrap [21], banana peel activated carbon [22] and orange peel [23]. Banana is one of the most consumed fruits in the world. Banana peel is mainly

used in composting, animal feeding and the production of proteins, methane, ethanol, pectin and enzymes. Its main constituents are cellulose, pectin, chlorophyll and other low molecular weight species. Carboxyl, hydroxyl and amide groups existing on the banana peel surface play a critical role in the adsorption processes [24]. Minerals like K, Mg, Na, Ca and Fe are mainly present in the banana peel. Banana peel is a lignocellulosic and low-cost environment friendly biomaterial that is readily available in large quantities. Rhodamine-B is carcinogenic, has neurotoxic effect and if ingested by human or animal bodies causes eyes and skin irritation and serious respiratory tract problems [25]. The present study gives a method to prepare a value added product from one of the agricultural wastes and present a cost-effective alternative to existing commercially available adsorbents.

EXPERIMENTAL

In the present study methyl orange, rhodamine-B, sulfuric acid, sodium hydroxide and hydrochloric acid were purchased from Merck. All other chemicals used were of Analar grade or chemically pure substances and used without further purification.

Synthesis of activated banana peel carbon from banana peels: Banana peels were obtained from the local market of Prayagraj city, India. Firstly, the peels were washed with distilled water to remove any adhering impurities and then dried under sunlight for 48 h. Dried banana peels were burnt to ash in the porcelain dish. The ash, thus obtained, was activated with H_2SO_4 and the excess acid was removed by repeated washing with distilled water. Washed and activated black colored carbon was left overnight for drying.

Procedure: Different concentrations of solutions of methyl orange and rhodamine-B were prepared by diluting the stock solutions with double distilled water. Requisite amount of activated banana peel carbon was added to the dye solution of desired concentration. After stirring for a preselected time, the solution was filtered (Whatman No. 1) and the remaining amount of dyes in the filtrate was measured spectrophotometrically by measuring the absorbance at 463 nm [26] and 554 nm [27], respectively for methyl orange and rhodamine-B. Effects of duration of treatment, amount of banana peel activated ash, pH and initial concentration on the removal of dye were studied by changing the variables one by one keeping other factors constant. This process was performed to determine the optimum conditions under which maximum removal of methyl orange and rhodamine-B could be obtained. All the experiments were conducted at room temperature using 1 g of carbon activated with 1.5 mL of concentrated sulfuric acid. Percentage removal of dye has been calculated by the following equation:

$$\text{Removal of contaminant } (\%) = \frac{C_i - C_f}{C_i} \times 100$$

where, C_i = initial concentration of contaminant and C_f = final concentration of contaminant

Instrumentation: Remaining amount of methyl orange and rhodamine-B in the filtrate was measured with a double beam spectrophotometer (Systronics 2203), pH of the solution was maintained with standard solutions of NaOH (0.1N) and HCl (0.1N). For measuring pH and temperature of the solution a digital pH meter (μ-pH System 361, Systronics) was used. FTIR spectra were recorded on a spectrum 2 Perkin-Elmer Spectrophotometer version 10.4.00 FTIR spectrophotometer. Scanning electron microscope (SEM) analysis was carried out using on JEOL (JSM 640 LV) scanning electron microscope equipped for energy dispersive analysis by X-ray (EDAX) after samples had been coated in platinum to investigate the morphological changes in banana peel carbon before and after activated with sulfuric acid. Powder X-ray diffraction (PXRD) analysis was carried out using an Rigaku smartlab 3KW to obtain structural information.

RESULTS AND DISCUSSION

Characteriazation of adsorbent

PXRD analysis: Before and after activation the prepared banana peel carbon was firstly characterized with the help of X-ray diffraction (XRD) using Cu-$K\alpha$ radiation. Presence of distinguished peaks in the XRD patterns before activation (Fig. 1a) clearly indicate the crystalline nature of material. Broad diffraction peaks obtained in the XRD pattern after activation (Fig. 1b) showed a amorphous nature of the material. Presence of a broad peak in the range of 20-30° demonstrates typical amorphous nature of amorphous carbon [28]. Moreover, an absence of sharp peaks indicates the negligible existence of residual carbon in the structure.
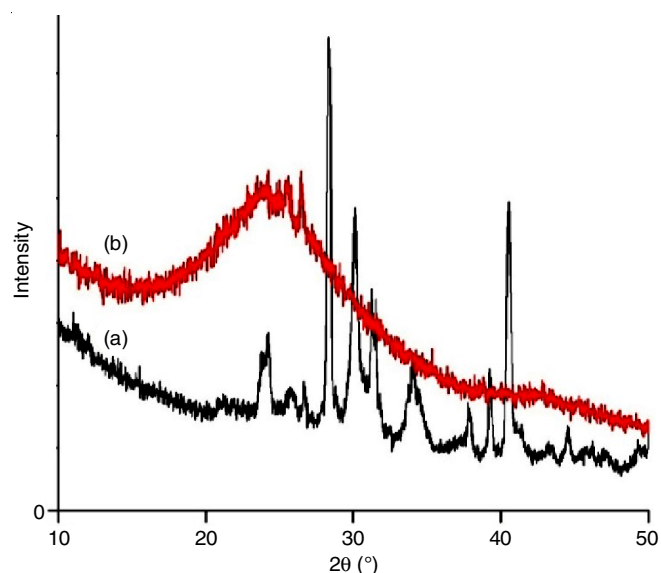

Fig. 1. PXRD patterns of banana peel carbon before (a) and after (b) activation

SEM analysis: Surface morphology of banana peel carbon (BPC) was studied by taking SEM images of the ash before activation, after activation and after removal of the dyes. Fig. 2a shows SEM image of BPC before activation in which particles having irregular shape and heterogonous nature indicate the amorphous structure of material [29,30]. Pores on the surface support the adsorption process. Fig. 2c shows SEM image of BPC after activation, while SEM images of BPC after adsorption of methyl orange and rhodamine-B, respectively are shown in Fig. 2e and 2g. Appearance of rough surface having carter-like pores partially covered by the dye can be clearly seen. It is also cleared that the pores become thicker in size and blocked after adsorption of dyes in the case of methyl orange.
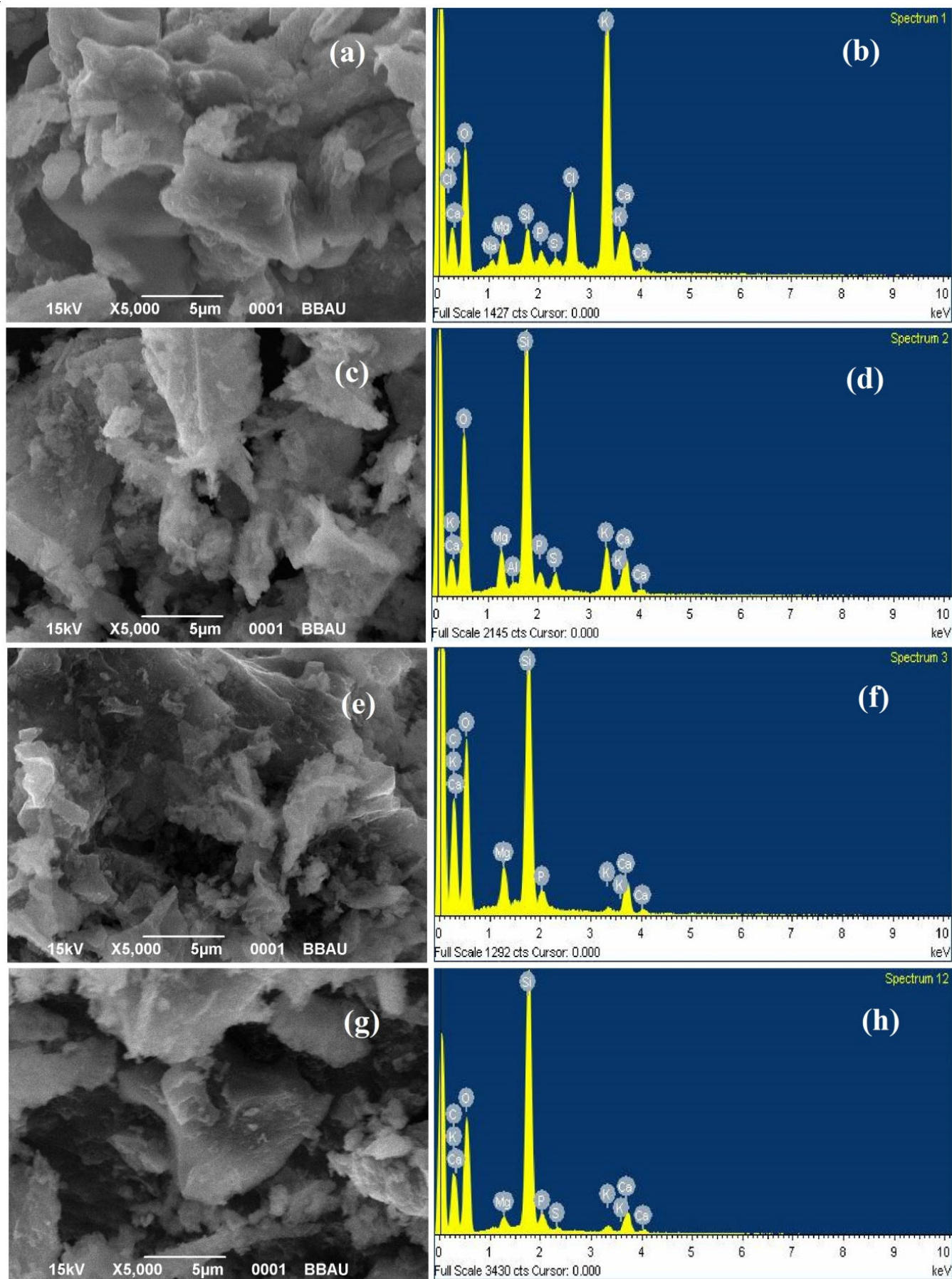
Fig. 2. SEM & EDX images: (a) & (b) BPC; (c) & (d) BPC after activation; (e) & (f) BPC after removal of methyl orange; (g) & (h) BPC after removal of rhodamine-B

SEM image of BPC loaded with rhodamine-B (Fig. 2g) clearly shows that the caves, pores and surfaces of adsorbent are covered by dye and the surface becomes smooth. It is evident that the adsorbent surface changes after adsorption of rhodamine-B. The EDX analyses of banana peel carbon before activation and after activation are shown in Fig. 2b and 2d. EDX analyses of ash after removal of methyl orange and rhodamine-B after removal are shown in Fig. 2f and 2h). EDX analyses after removal of dyes strongly support the adsorption of methyl orange and rhodamine-B dyes. Elemental compositions of carbon before and after activation indicate the presence of K, Ca, P, Si, Cl as the major elements followed by Mg, O and S as the minor element in banana peel carbon [31], appearance of oxygen is clearly observed in the EDX analysis of carbon after adsorption of dyes.

FTIR analysis: The FTIR spectra of methyl orange dye adsorbed on banana peel carbon (BPC) before and after activation are shown in Fig. 3. A peak at 3439 cm^{-1} is attributed to the stretching vibration of free and intermolecular bonded -OH groups or amine (-NH) groups. Small peak at 2923 cm^{-1} is attributed to stretching vibration of -CH group. The peaks at 1624 and 1468 cm^{-1} can be attributed to stretching vibration of carboxyl group. Sharp bands observed at 1077 and 1042 cm^{-1} can be attributed to C-O stretching vibration in alcoholic group and C-N stretching vibrations, respectively, thus showing the adsorption of dye on sorbent surface. In the FTIR spectra of banana peel carbon after activation and after loading of methyl orange, the appearance of new bands at 1385, 805, 593 cm^{-1} and disappearance of band at 1441 cm^{-1} along with shifting of the bands from 2931 to 2919 cm^{-1}; 2855 to 2851 cm^{-1}; 1627 to 1636 cm^{-1}; 601 to 665 cm^{-1} may be due to the involvement of some functional group present on methyl orange dye through weak electrostatic interaction or van der Waals forces [32]. Adsorption of rhodamine-B dye on banana peel activated carbon was confined by new peaks obtained at 2070.5 and 1741.20 cm^{-1}, which are related to =C-H and -COOH aromatic groups, respectively of rhodamine-B. Shifting of other peaks suggested that the synthesized activated banana peel carbon has interacted with rhodamine-B dye [33,34].
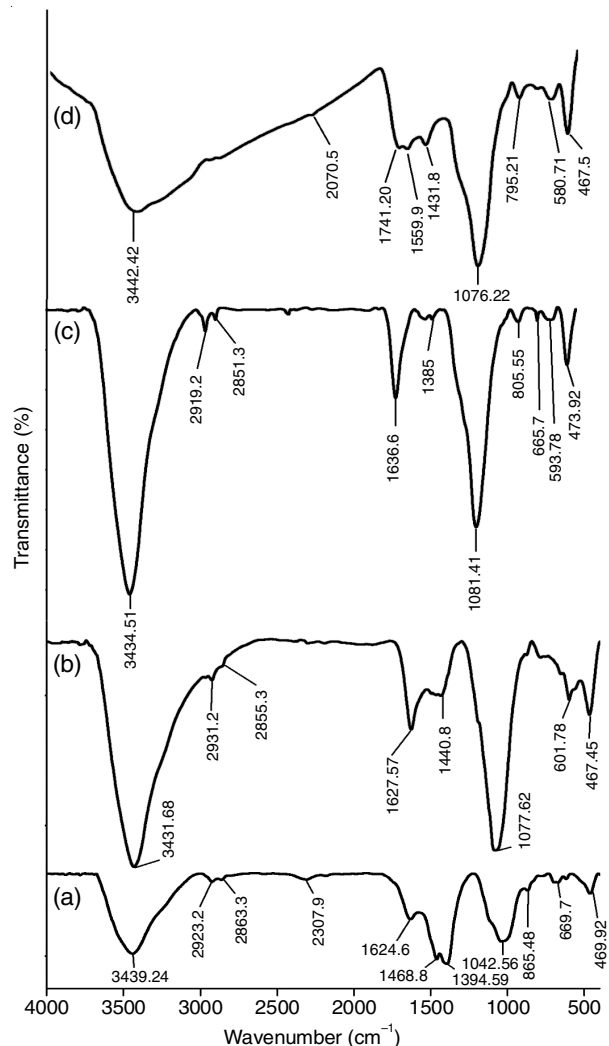

Fig. 3.   FTIR spectra of (a) BPC (b) BPC after activation (c) BPC after removal of methyl orange and (d) BPC after removal of rhodamine-B

Effect of time: Under particular conditions to find out the minimum time required for the maximum removal of dyes, the duration of treatment was changed keeping the other conditions constant. Results given in Table-1 (entries 1-5 and Fig. 4)

TABLE-1
OPTIMUM CONDITIONS FOR THE REMOVAL OF METHYL ORANGE AND RHODAMINE-B CONTAMINATED FROM WATER

Time (min)	pH	Concentration (ppm)	Amount of adsorbent (mg)		Removal (%)	
			Methyl orange	Rhodamine-B	Methyl orange	Rhodamine-B
30	5.6	10	100	125	80.0	88.0
45	5.6	10	100	125	85.2	96.3
60	5.6	10	100	125	91.1	97.5
75	5.6	10	100	125	98.5	99.0
90	5.6	10	100	125	98.1	98.5
75	2.5	10	100	125	98.3	98.8
75	4.5	10	100	125	98.3	98.8
75	7.5	10	100	125	88.2	88.5
75	10.5	10	100	125	77.3	68.3
75	5.6	02	100	125	98.4	98.9
75	5.6	05	100	125	98.4	98.9
75	5.6	15	100	125	87.8	92.2
75	5.6	20	100	125	76.7	70.0
75	5.6	10	25	50	41.5	86.0
75	5.6	10	50	75	70.2	91.2
75	5.6	10	75	100	83.0	96.1
75	5.6	10	125	150	98.0	98.5

Fig. 4.   Effect of variation of time on the removal of methyl orange and rhodamine-B
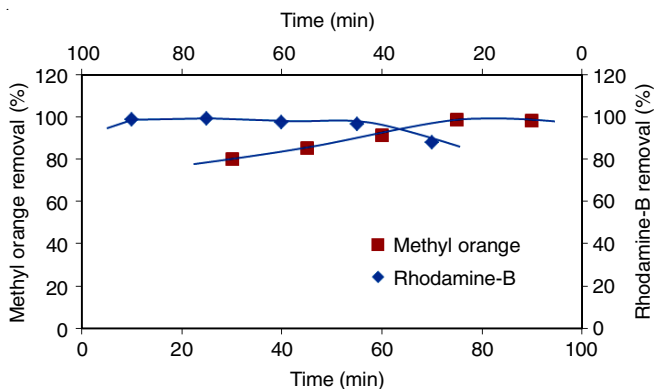

Fig. 6.   Effect of variation of concentration on the removal of methyl orange and rhodamine-B

clearly showed that maximum removal was obtained in the beginning itself and further stirring for the long duration starts decreasing the percentage removal of methyl orange and rhodamine-B.

Effect of pH: pH of the medium being the most important variable thus study was performed to investigate the effect of the variation of pH of contaminated water on the efficiency of dye removal. To find out the optimum conditions for the maximum removal of dyes, pH of the dye solution was changed (from 2.5 to 10.6), while keeping other variables constant. It was observed that removal efficiency of the activated carbon remains almost constant in a wide range of pH (from 2.5 to 5.6) but on further increasing the pH above 5.6, the removal efficiency starts decreasing (Table-1; entries 4, 6 to 9 and Fig. 5). This suggests that acidic conditions were more favourable for the removal of dyes. Probably H^+ ions of the solution were involved in the reduction of dyes [35,36]. This is a very important finding from the industrial point of view because in most of the reported studies optimum efficiency is obtained only in a limited range of pH [37].
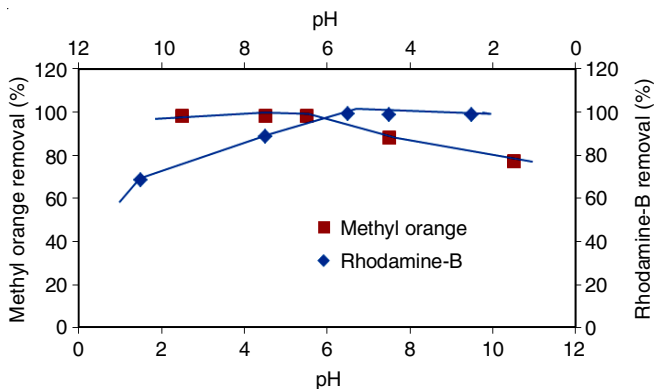

Fig. 5.   Effect of variation of pH on the removal of methyl orange and rhodamine-B

Effect of concentration: To find out maximum removal of methyl orange and rhodamine-B from the contaminated water the initial concentration of dyes solution was changed keeping the other conditions constant. It was observed (Table-1; entries 4, 10 to 13 and Fig. 6) that at room temperature from 25 mL aqueous solution containing 10 ppm dye, 100 mg of adsorbent was able to remove 98.5% dye in the case of methyl orange, while in the case of rhodamine-B 125 mg adsorbent
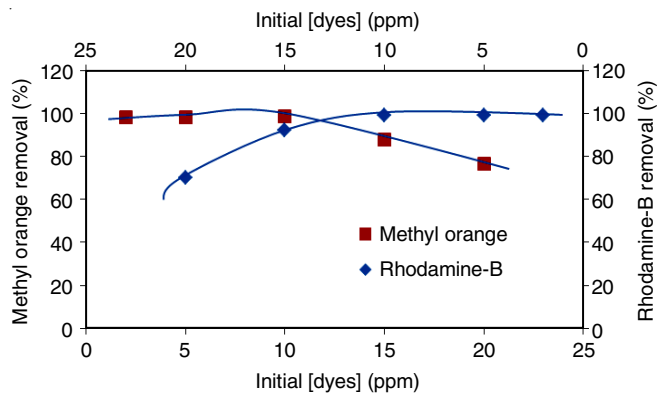
was able to remove 99 % dye in 75 min. It was also observed on further increasing the initial concentration of dye in solution, the percentage of removal decreased. This might be due to the fact that after saturation of active sites, excess dye remains in the solution, which decreases the percentage of removal.

Effect of adsorbent dose: Results of the amount variation of adsorbent on the percentage removal of methyl orange and rhodamine-B from contaminated water are given in (Table-1, entries 4, 14 to 17 and Fig. 7). It is clear that as expected an increase in the adsorbent amount, increases the removal efficiency and after that the efficiency of adsorbent to remove the dye becomes constant.
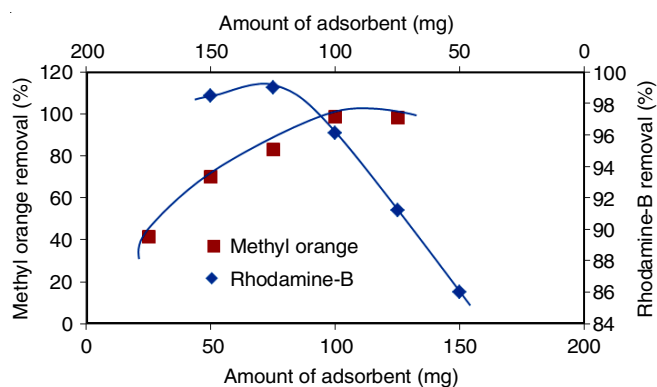

Fig. 7.   Effect of variation of amount of adsorbent on the removal of methyl orange and rhodamine-B

Conclusion

The present study gives a method to prepare a value added product from one of the agricultural wastes and presents a cost-effective alternative to existing commercially available adsorbents. It has been shown that an amorphous material obtained after the acid activation of carbon of banana peel is an effective adsorbent for removal of methyl orange and rhodamine-B dyes from aqueous solution. Activation of the carbon of banana peel was done simply with sulphuric acid and its preparation is extremely simple without the requirement of any functionalization, chemical treatments and the elevated temperature. A comparative study has carried as mentioned in Table-2, where various activated carbon prepared from agricultural wastes and used for dye removal. It is clear from Table-2 that best percentage removal of dye achieved from activated

TABLE-2
COMPARISON OF REMOVAL OF METHYL ORANGE AND RHODAMINE-B ONTO ACTIVATED CARBON

Rhodamine-B				Methyl orange			
Adsorbent	Time (min)	Removal (%)	Ref.	Adsorbent	Time (min)	Removal (%)	Ref.
Silk cotton carbon	60	4.1	[38]	Spent tea leaves carbon	63.8	58.2	[40]
Coconut tree saw dust carbon	60	55.0	[38]	Lemon peel carbon	60	50.0	[41]
Maize cob carbon	180	55.5	[38]	Sea mango carbon	60	80.0	[42]
Sago waste carbon	210	91.0	[39]	Banana peel carbon	75	98.5	This study
Banana peel carbon	75	99.0	This study	–	–	–	–

carbon from banana peel. At room temperature, 0.1 g of prepared material was able to remove 98.5 % anionic dye methyl orange and 99.0% cationic dye rhodamine-B from contaminated water having 10 ppm dye in 75 min. Efficiency of the system remains constant in a wide range of pH from 2.5 to 5.6.

CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this article.

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