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Optimization of brilliant green biosorption by native and acid-activated watermelon rind as low-cost adsorbent

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ABSTRACT

The use of low-cost and eco-friendly adsorbent such as watermelon rind (WR) has been investigated as an ideal alternate for removing dyes from wastewater. This study deals with the removal of textile dye brilliant green (BG) from aqueous solutions by native WR and chemically activated watermelon rind (AWR). Chemical activation of WR was carried out using 1:1 ortho-phosphoric acid. The loading capacity of WR and AWR was found to be 92.6 and 188.6 mg g^{-1}, respectively. The equilibrium data were fit to Langmuir, Freundlich, and Temkin isotherms and found to have better fit to Langmuir isotherm. The sorption of BG onto WR and AWR was rapid and follows pseudo-second-order kinetic model. Thermodynamic parameters including change in free energy (ΔG°), enthalpy (ΔH°), and entropy (ΔS°) are derived and found to be spontaneous and exothermic in nature. These observations suggest that WR can be used as a non-hazardous agro material for removal of BG from aqueous solutions.

Keywords: Watermelon rind; Activation; Brilliant green dye; Adsorption

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1. Introduction

Rapid increase in industrialization has resulted in the discharge of large amounts of colored dye effluents into water-receiving bodies [1,2]. Discharge of colored dye effluents into natural resources has caused many problems like carcinogenicity, skin allergy, and irritation. Brilliant green (BG) is one of the triarylmethane dye occurs as small, shiny, golden crystals soluble in water or alcohol used to dye silk and wool, and for the production of cover paper in the paper industry [3]. BG dye is toxic, mutagenic, and carcinogenic that affects aquatic life and humans [4]. Especially, it causes eye burn, which may be responsible for permanent injury to the eyes of humans and animals [5].

During the past few years, colored dye effluents are normally treated by physical and chemical treatment process which are less adaptable and expensive [6]. It is now well established that for the wastewater treatment, adsorption has several advantages, such as being cost effective, easy to operate, and rapid technique, over other methods [7,8]. Moreover, the ability of adsorption to remove toxic chemicals without producing any toxic byproducts, thereby keeping the quality of water undisturbed, has also popularized the adsorption technique in comparison with other techniques [9]. Activated carbon is found to be the most effective adsorbent for the removal of dyes from

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industrial effluents. But the cost of activation and difficulty in regeneration of spent activated carbon has encouraged researchers to look for alternative adsorbents for the removal of dyes from industrial effluents [10,11]. Biosorption is a "cost-effective technique" for dye removal [12–14]. Many low-cost agricultural wastes, such as peanut husk [15], rice husk [16], coconut husk [17], rejected tea [18], and fruit peels, such as banana peel, orange peel [19], jackfruit peel [20], custard apple shell [21], and pomegranate peel [22], are used as sorbents for the removal of dyes. The disadvantages of adsorption technique by low-cost adsorbents are low stability and low removal efficiency. The stability and removal efficiency of raw biomass can be further improved by chemical modification and activation. In view of this, several agricultural wastes have been chemically modified and proven to be better sorbents for the removal of dyes. In continuation to these efforts, watermelon rind (WR) was chemically activated with ortho-phosphoric acid and evaluated for the removal of BG in native and activated forms.

Watermelon (*Citrullus lanatus*), being the largest and heaviest fruit, is one of the most abundant and cheap fruits available in India. Watermelon production occupies 6–7% of overall fruit production and is high during summer because of its tropical nature. WR is a byproduct of watermelon, which consists of many constituents like proteins, pectin, cetrulline, and careteniods [23–25]. Due to the presence of hydroxyl, corbonyl, and carboxyl groups, the WR is known to bind cations in aqueous solutions [26,27]. This study was undertaken to evaluate the application potential of WR as an inexpensive and eco-friendly adsorbent for the removal of BG from water. Effect of parameters like pH, adsorbent dose, contact time, initial dye concentrations, and temperature were investigated for the sorption of dyes. Finally, isotherms, thermodynamic, and kinetics, as well as the diffusion parameters were evaluated for the experimental data.

2. Materials and methods

2.1. Preparation of adsorbent

WRs were obtained from local fruit market and washed under tap water several times, followed by washing double-distilled water. After thorough washing, the WR was cut into small pieces and dried under sunlight for 7 d to remove all moisture content present. Later the dried WR pieces were washed with hot water (70˚C) to remove any soluble matter present and dried in oven at 85˚C for 48 h. The oven-dried WR was powdered using conventional mixture and sieved through 100 mesh range. The sieved WR powder was stored in desiccators and used for batch experiments. Activated watermelon rind (AWR) was prepared by adding ortho-phosphoric acid in 1:1 ratio and activated in oven at 110˚C for 4 h. The obtained char was washed with double-distilled water to remove excess acid present and dried in oven at 110˚C for 2 h. Then, the oven-dried AWR was powdered using conventional mixture and sieved through 0.150–0.080 mm (100 BSS mesh) sieve. The sieved WR and AWR were used for sorption studies.

2.2. Preparation of dye solutions

The 1,000 mg L^{-1} stock solution was prepared by dissolving 1 g of BG in 1 L of deionized water. pH adjustments were done using 0.1 M HCl and 0.1 M NaOH solutions. All reagents were of AR grade and deionized water was used for dilutions. The chemical structures of BG are represented in Fig. 1.

2.3. Adsorption studies

Batch experiments were conducted at room temperature in a roto-spin unit at 50 rpm using 50-ml Tarsons tubes. The effect of adsorption parameters, such as adsorbent dosage, contact time, pH initial dye concentration, salt ionic strength, and temperature,

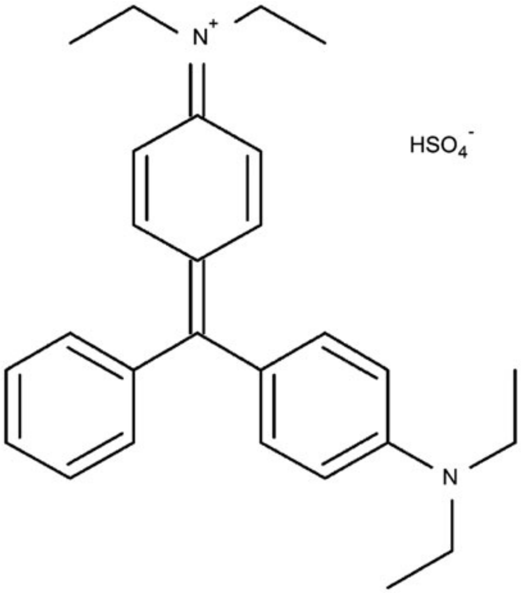

Fig. 1. Chemical structure of BG dye.

were studied. The effect of pH was evaluated by varying the pH from 4 to 10 using 0.1 M HCl and 0.1 M NaOH for adjustments. The contact time was varied between 10 and 120 min to study the effect of time on sorption and the adsorbent dosage was varied from 0.5 to 5 g L^{-1} to know the effect of dosage, while the rest of the parameters were kept constant for each parameter studies. The sorption capacity of BG was determined by contacting 1.5 g L^{-1} of WR and AWR with 20 ml of known concentration of dye solution (50–300 mg L^{-1}). For each experiment, the solid phase was separated with centrifugation at 5,000 rpm for 15 min and residual concentration present in the supernatant was determined by UV–vis spectrophotometer at 623 nm.

2.4. Instrumentation techniques

UV–vis spectrophotometer (Hitachi U-2800 spectrophotometer) was used to determine the concentration of dyes. FTIR Spectroscopy (Thermo Nicolet, AVATAR 330) was used to know the functional groups present in the WR. The FTIR spectra of WR and AWR, and metal-loaded WR and AWR were recorded in mid-IR region in the range of 4,000–400 cm^{-1} by KBr pellet method.

3 Results and discussions

3.1. Characterization of WR and AWR

The sorption of dye ions onto plant materials is attributable to the active groups and bonds present on them. In order to identify the major functional groups present in WR and AWR, preliminary qualitative analysis was done with FTIR spectroscopy (Fig. 2). The FTIR spectra of WR displayed a number of peaks pertaining to different functional groups. The broad and intense peak around 3,371 cm^{-1} corresponds to –OH stretching vibrations of cellulose, pectin, and lignin. The peak at 2,917 cm^{-1} is attributed to –CH stretching vibrations of methyl and methoxy groups. The peak at 1,734 cm^{-1} corresponds to –C=O stretching of carboxylic acid or esters, and asymmetric and symmetric vibrations of ionic carboxylic groups ($–COO^{-}$), respectively, appeared at 1,633 and 1,423 cm^{-1} [28]. The peak at 1,383 cm^{-1} is attributed to symmetric stretching of $–COO^{-}$ of pectin [29]. The peaks from 1,350 to 1,000 cm^{-1} can be attributed to stretching vibrations of carboxylic acids and alcohols. The FTIR spectrum of AWR displayed similar kind of peaks compared to that of WR. It is well indicated from the FTIR spectrums of WR and AWR that carboxylic and hydroxyl groups are abundantly present and as biopolymers these groups act as proton donors. The comparisons of FTIR spectra of native, activated, and dye-loaded WRs are presented in Table 1. The shifts in peaks for dye-loaded AWR may be attributed to changes in counter ions associated with carboxylate and the hydroxylate anions suggesting that acidic, carboxyl, and hydroxyl groups are predominant contributors in dye removal. The removal process can be attributed to negatively charged carboxylate and hydroxylate anions of AWR that can electrostatically interact with the positive charges of dyes.

Scanning electron microscope analysis was performed in order to study the morphology of the native and acid-activated WR. The surfaces of both WR and AWR were found to be porous and no significant changes was observed (Fig. 3).

The point zero charge (pH_{pzc}) of the WR and AWR was determined by solid addition method (pH drift method). To a series of 50-ml Tarsons tubes, 30 ml of 0.1 M KCl solution was added and initial pH_i was adjusted from 2 to 10 by 0.1 M HCl or 0.1 M NaOH. To the 30-ml solution, 0.1 g of WR sorbent was added to each of the tube. The suspensions were then shaken manually and allowed to equilibrate for 48 h. After 48 h, the suspension was filtered and the pH_f values of the supernatant liquid were noted. Similar experiments were performed for the AWR sorbent. The difference between the initial and final pH values (ΔH) was plotted against pH_i (Fig. 4). The pH_{pzc} values for WR and AWR were experimentally found to be pH 4.9 and 4.1, respectively.

3.2. Batch adsorption studies

3.2.1. Effect of pH

One of the important parameters considered during adsorption is pH, due to competitive adsorption for active sites by H^{+} ions. The effect of pH was studied by varying the pH in the range of 4–10 for BG. Since BG decolorizes at low pH due to the structural changes, the effect of pH was studied from pH 4–10 (Fig. 5). It was observed that removal efficiency of BG increased with increase in pH for WR and found to be maximum at pH 7. But in the case of AWR, pH has no effect on the removal of BG from aqueous solution. This indicates that the electrostatic mechanism was not the only mechanism for BG adsorption by AWR. Hence, further experiments were carried out at pH 7 for WR and at pH 5 (native pH of BG in aqueous solution) for AWR.

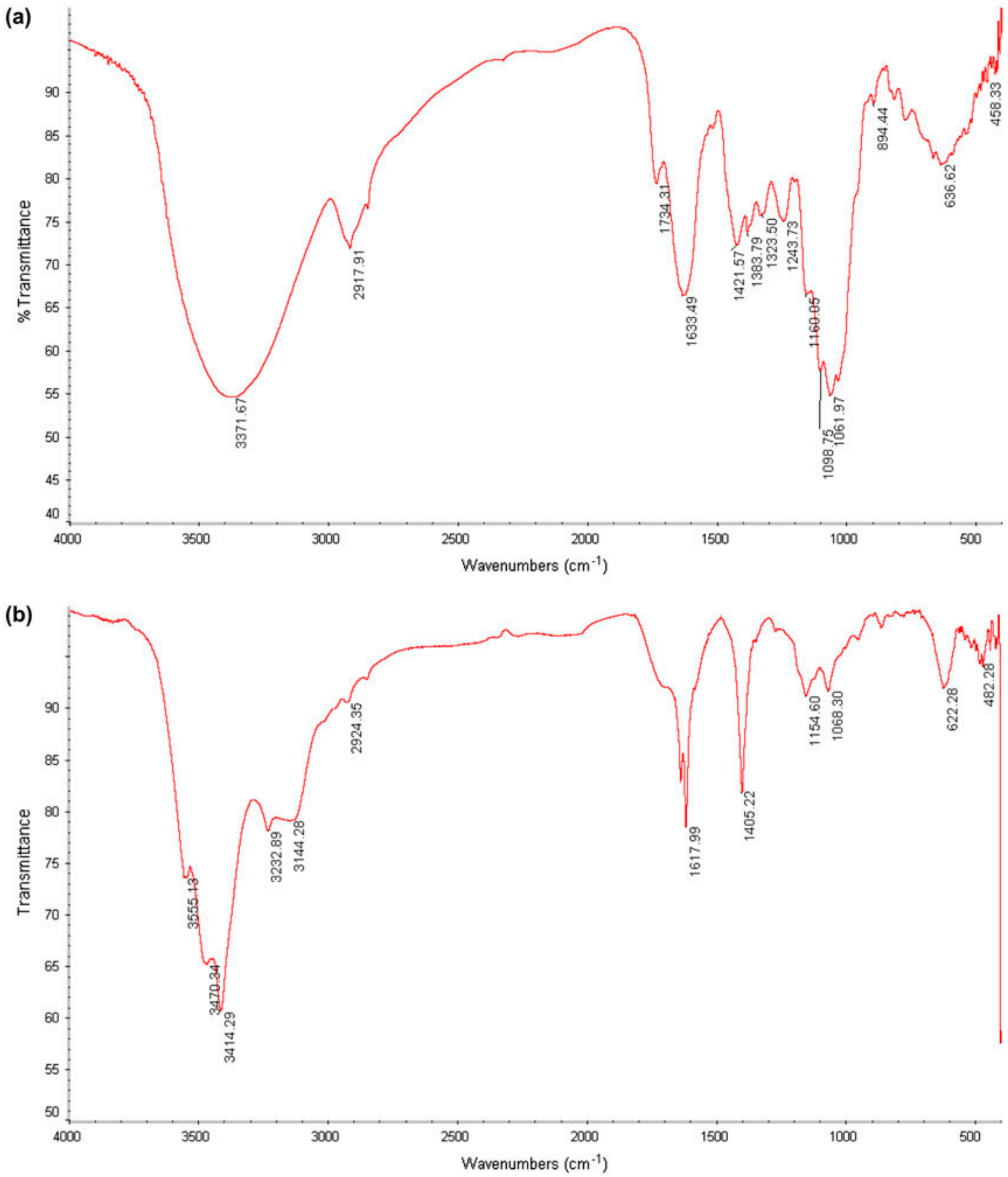

Fig. 2. FTIR spectra of (a) native and (b) acid-treated WR.

3.2.2. *Effect of adsorbent dosage*

Adsorbent dosage is one of the important parameter studied when conducting batch mode studies. The effect of adsorbent dosage on the removal of BG was studied by varying the dosage from 0.5 to 5 g L^{-1} (Fig. 6). Optimal removal efficiency of BG was observed at 1.5 g L^{-1} dose of WR and AWR. Increase in adsorption by increase in adsorbent dose is due to the availability of more number of active sites on the surface and the decrease in efficiency at higher concentration of adsorbent is due to the decrease in surface active sites as a consequence of partial aggregation of adsorbent.

3.2.3. *Effect of salt ionic strength*

It is important to discuss the effect of salt ionic strength on the adsorption of cationic dyes onto WR and AWR. Industrial dyeing effluents usually contain

Table 1
Wave number (cm^{-1}) of FTIR peaks of different WR samples

Functional groups	WR	BG-loaded WR	AWR	BG-loaded AWR
–OH stretching vibrations of cellulose	3,371	3,417	3,414	3,312
–CH stretching vibrations of methyl groups	2,917	2,939	2,924	2,918
–C=O stretching of carboxylic acid or esters	1,734	1,730	1,712	1,720
$–COO^-$ asymmetric vibrations of ionic carboxylic groups	1,633	1,620	1,618	1,610
$–COO^-$ symmetric vibrations of ionic carboxylic groups	1,423	1,402	1,405	1,403

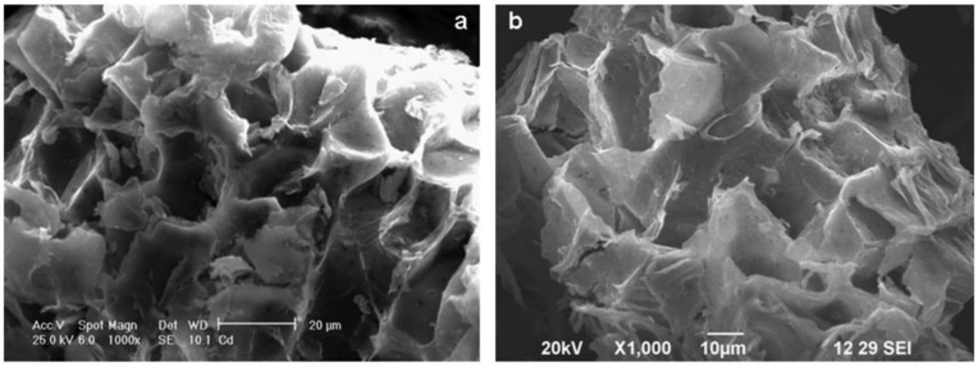

Fig. 3. SEM image of (a) native and (b) activated WR.

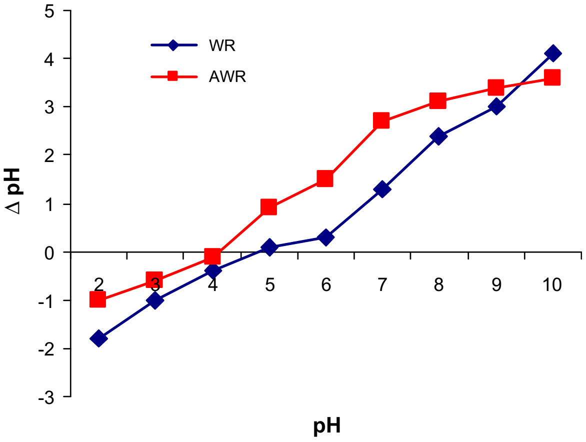

Fig. 4. Plots of point zero charge (pH_{pzc}) of native and acid-treated WR.

high salt concentration. In order to study the adsorption capability of WR and AWR towards cationic dyes in the presence of ionic salts, salts such as NaCl and $CaCl_2$, with different ionic strengths were used to stimulate the salt ionic in water. The results are summarized in Table 2. From Table 2, it is evident that the presence of ionic salts in the solution has greatly influenced the sorption capacity of WR, but in the case of AWR the effect is minimal. The substantial decrease in the sorption capacity observed for WR can be attributed to competitive adsorption for active sites between Na^+ and Ca^{2+} ions and positively charged dye ions. The minimal effect shown by salt ionic strength on sorption of BG by AWR is mainly due to surface pore adsorption and not based on electrostatic attraction. If the electrostatic attraction is the only mechanism operated, then the presence of salt ionic strength will have some effect due to competitive adsorption. These results can be corroborated with the effect of pH on the adsorption of BG by WR and AWR.

3.2.4. Desorption and regeneration studies

Desorption and regeneration studies were performed to know the reusability potential of WR and AWR. Desorption of dyes from WR and AWR is studied by using 0.1 M HCl, 0.1 M acetic acid, distilled water, and 0.1 M NaOH as desorbing agents. About 0.05 g of dye-loaded WR and AWR was contacted individually with 20 ml of desorbing agents for 30 min at room temperature in an orbital shaker at 150 rpm. The desorbed supernatant solution was subjected to UV–vis spectrophotometer for residual concentration of dyes. It was found that 0.1 M acetic acid has the highest desorbing capacity for WR, followed by 0.1 M HCl,

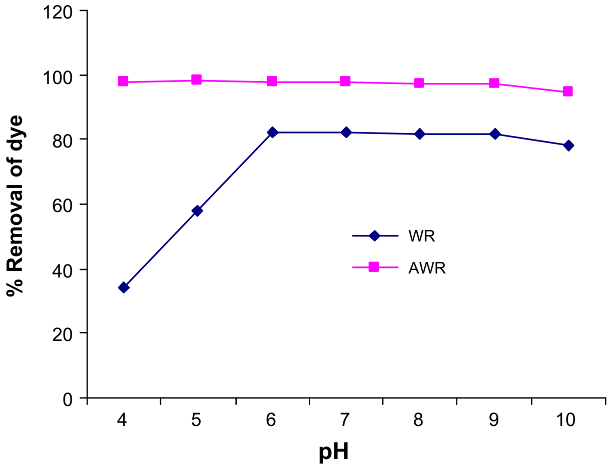

Fig. 5. Effect of pH on removal of BG by WR and AWR from aqueous solution (initial concentration 50 mg L^{-1}, dose 1.5 g L^{-1}, time 60 min, and temperature 303 K).

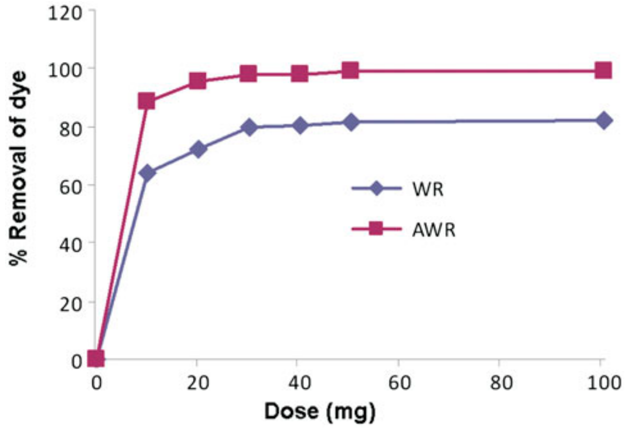

Fig. 6. Effect of adsorbent dose on removal of BG from aqueous solution (pH 5 and 7 for WR and AWR, initial concentration 50 mg L^{-1}, time 30 min, and temperature 303 K).

distilled water, and 0.1 M NaOH (Fig. 7). But in the case of AWR, acetic acid and HCl showed the highest desorption, followed by 0.1 M NaOH and H_2O. From these results, it is can be said that the desorption and reusability potential of AWR are high compared to that of WR. Hence, AWR can be an effective adsorbent for removal of BG from industrial effluents.

3.3. Adsorption kinetic studies

In order to study the kinetics of adsorption, the removal of BG by WR and AWR was taken at preset time intervals and concentrations of dyes were similarly measured. It was observed that the removal of BG by WR and AWR was rapid in first 10 min and reached equilibrium within 30 min. The rapid removal in the first 10 min is due to immediate availability of more number of active sites at the surface for adsorption. To examine the rate-controlling mechanism of the present process, the experimental data were fit to well-known kinetic models, such as pseudo-first-order, pseudo-second-order, and intraparticle diffusion models.

Table 2
Effect of salt ionic strength on removal of BR from aqueous solution by WR and AWR

Salt strength (mol L^{-1})	Salt	Loading capacity (mg g^{-1}) WR	AWR
0	–	23.3	32.1
0.1	NaCl	17.6	29.7
	$CaCl_2$	16.4	29.3
0.2	NaCl	12.5	28.4
	$CaCl_2$	12.4	27.9
0.3	NaCl	7.8	27.7
	$CaCl_2$	6.9	26.6

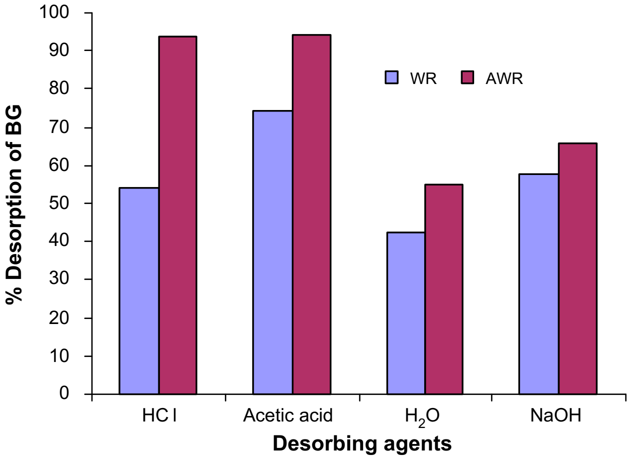

Fig. 7. Desorption and regeneration studies on removal of BG by WR and AWR (dose 2.5 g L^{-1}, time 30 min, and temperature 303 K).

The pseudo-first-order rate equation of Lagergren is represented as:

$$\ln (q_e - q_t) = \ln q_e - k_1 t \tag{1}$$

Table 3
Kinetic parameters derived for removal of BG from aqueous solution by WR and AWR (pH 5 and 7 for WR and AWR, initial concentration 50 mg L^{-1}, dose 1.5 g L^{-1}, and temperature 303 K)

		Pseudo-first-order constants			Pseudo-second-order constants		
Sorbent	Experimental q_e (mg g^{-1})	q_e (mg g^{-1})	k_1 (min^{-1})	R^2	q_e (mg g^{-1})	k_2 (g mg^{-1} min^{-1})	R^2
WR	23.3	7.442	0.027	0.903	22.7	0.035	0.999
AWR	32.1	4.794	0.007	0.756	31.5	0.097	0.999

where q_e is the amount of metal adsorbed at equilibrium (mg g^{-1}), q_t is the amount of metal adsorbed at time *t*, and k_1 is the first-order reaction rate constant. A straight line of ln $(q_e - q_t)$ vs. *t* suggests the applicability of this kinetic model, and the values of k_1 and q_e were determined from the plot.

Based on the sorption equilibrium capacity the pseudo-second-order equation can be expressed as:

$$\frac{t}{q_t} = \frac{1}{K_2 q_e^2} + \frac{t}{q_e} \tag{2}$$

The respective parameters of pseudo-first-order and pseudo-second-order kinetic models for removal of BG from aqueous solution by WR and AWR are summarized in Table 3. It was observed that correlation coefficients of pseudo-first-order kinetic model were low and the theoretical q_e values are also not close to the experimental values. It was also observed that the experimental data fit better to the pseudo-second-order kinetic model. The better fit is further supported by its correlation coefficients. The theoretical q_e values were also close to the experimental values. These observations suggest that the removal of BR by WR and AWR from aqueous solution follows the pseudo-second-order reaction. According to the pseudo-second-order model, the boundary layer resistance is not the rate-limiting step, the external resistance model cannot adequately describe the adsorption mechanism, and the process controlling the rate may be a chemical sorption involving valence forces through sharing or exchanging of electrons between sorbate and sorbent.

The kinetic data were further analyzed using intraparticle diffusion model in order to study the steps of diffusion mechanisms.

$$q_t = k_{int} t^{1/2} + C \tag{3}$$

A plot of q_t vs. $t^{1/2}$ should result in a straight line if the adsorption mechanism follows intraparticle diffusion process only. If the plot shows multi-linear plots, it indicates that two or more steps take place. The plots of the present process resulted in two linear plots for WR and a straight line for AWR (Fig. 8). The linear plot of AWR suggests that the removal of BG follows intraparticle diffusion process. The first linear plot of BG for WR is due to the immediate utilization of ample active sites on the adsorbent surface and the second linear plot is attributed to very slow diffusion of the adsorbate from the surface site into the inner pores [30]. Thus, the initial adsorption of BG by WR may be governed by intraparticle transport of surface diffusion and the later part may be controlled by pore diffusion [31]. However, the intercept of the line for all the studied dyes fails to pass through the origin which may be attributed to the difference in the rate of mass transfer in the initial and final stages of adsorption [32].

3.4. Adsorption isotherms

At equilibrium, WR and AWR were individually allowed to come into contact with varying concentrations (50–300 ppm) of BG. The maximum loading

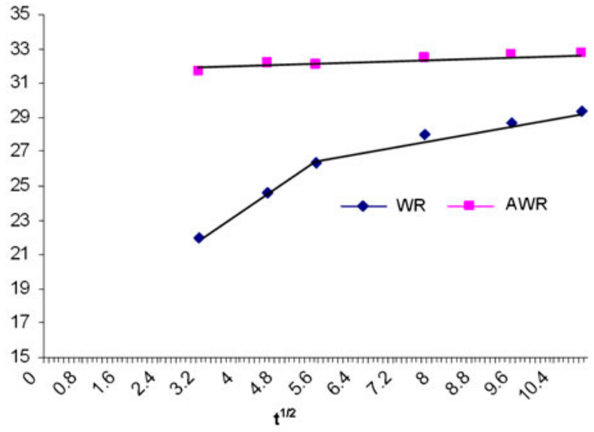

Fig. 8. Weber and Morris intraparticle diffusion plots for removal of BG by WR and AWR from aqueous solution (initial concentration 50 mg L^{-1}, dose 1.5 g L^{-1}, pH 7 for WR and pH 5 for AWR, and temperature 303 K).

capacity of WR and AWR was observed to increase with increase in initial concentration of dye. The maximum loading capacity of WR and AWR towards BG was found to be 92.3 and 188.9 mg g^{-1}, respectively (Fig. 9). In order to examine the relationship between the concentration of dye at equilibrium (C_e) and loading capacity (q_e) equilibrium data was analyzed with Freundlich, Langmuir, and Temkin isotherm models.

The linear form of Freundlich isotherm is given as:

$$\log q_e = \log K_f + \frac{1}{n}\log C_e \tag{4}$$

where K_f and n are Freundlich constants indicating adsorption capacity and intensity, respectively. If the E_q applies, a plot of log q_e vs. log C_e will be straight. The linear form of Langmuir equation after rearrangement is given as:

$$\frac{C_e}{q_e} = \frac{1}{bV_m} + \frac{C_e}{V_m} \tag{5}$$

where C_e is the concentration of solution at equilibrium (mg^{-1}), q_e is the amount of dye adsorbed per mass unit of adsorbent (mg g^{-1}), V_m is the amount of adsorbate at complete monolayer coverage (mg g^{-1}), and b is a constant that relates to the heat of adsorption (L mg^{-1}). If the biosorption follows Langmuir isotherm, then a plot of C_e/q_e should be a straight line with slope $1/V_m$ and intercept $1/bV_m$.

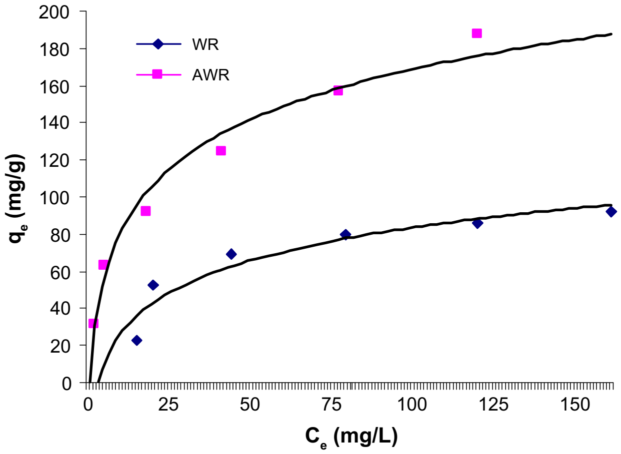

Fig. 9. Equilibrium plot of C_e vs. q_e for removal of BG from aqueous solution by WR and AWR (pH 5 and 7 for WR and AWR, dose 1.5 g L^{-1}, time 30 min, and temperature 303 K).

The Temkin isotherm considers the effect of adsorbate interaction on adsorption at active sites. The linear form of equation is given as:

$$q_e = B\ln A + B\ln C_e \tag{6}$$

where A is the equilibrium binding constant (L/mg) and B is related to the heat of adsorption. A plot of q_e vs. ln C_e enables the determination of isotherm constants.

The results of the Freundlich, Langmuir, and Temkin isotherm constants are represented in Table 4. The correlation coefficients of Freundlich and Temkin isotherms were low, which suggests that these models might not be applicable to the present system. The correlation coefficient of the Langmuir isotherm were very close to 1, which suggests that the Langmuir model fits better for adsorption of BG. The theoretical monolayer coverage (V_m) of BG on WR and AWR was calculated. Based on the Langmuir monolayer coverage, the loading capacity of WR and AWR was found to be higher than many other agricultural wastes reported in literature (Table 5).

Table 4
Isothermal parameters derived for removal of BG from aqueous solution by WR and AWR (pH 5 and 7 for WR and AWR, time 30 min, dose 1.5 g L^{-1}, and temperature 303 K)

Isotherm model	Parameters	WR	AWR
Freundlich	K_f	8.12	15.13
	$1/n$	0.075	0.048
	R^2	0.740	0.988
Langmuir	q_{max} (mg g^{-1})	95.2	192.3
	b (L mg^{-1})	0.091	0.062
	R^2	0.984	0.991
Temkin	A	35.98	35.01
	B	0.15	0.39
	R^2	0.816	0.906

Table 5
Comparison of WR and AWR sorbents with some previously reported literature

Adsorbent	q_e (mg g^{-1})	Reference
Rice husk ash	21.6	[33]
Bagasse fly ash	116	[34]
Bottom ash	28.9	[5]
Acorn	2.01	[3]
WR	92.6	This study
AWR	188.6	This study

Table 6

Thermodynamic parameters derived for removal of BG by WR and AWR from aqueous solution (pH 5 and 7 for WR and AWR, time 30 min, dose 1.5 g L^{-1}, and initial dye concentration 50 mg L^{-1})

Adsorbent	Temperature (K)	Adsorption capacity (q_e, mg g^{-1})	ΔG° (kJ mol^{-1})	ΔH° (kJ mol^{-1})	ΔS°(J mol^{-1} K^{-1})
WR	303	23.3	−1.103	−1.195	−90.3
	313	23.0	−1.020		
	323	22.6	−0.922		
AWR	303	32.1	−6.984	−5.943	−1,302
	313	32.6	−9.071		
	323	32.7	−9.588		

3.5. Thermodynamics of adsorption

In order to describe the thermodynamic behavior of sorption of BG onto WR and AWR at equilibrium, the temperature was varied from 303 to 323 K. The thermodynamic parameters including change in free energy (ΔG°), enthalpy (ΔH°), and entropy (ΔS°) were derived from the following equations:

$$K_D = \frac{q_e}{C_e} \tag{7}$$

$$\Delta G^\circ = -RT \ln K_D \tag{8}$$

$$\Delta G^\circ = \Delta H^\circ - T\Delta S^\circ \tag{9}$$

where K_D is the equilibrium constant related to the Langmuir constant "*b*", R is the universal gas constant (8.314 J mol^{-1} K^{-1}), and T is temperature in Kelvin.

The thermodynamics parameters calculated for the present systems are presented in Table 6. It can be observed from the table that the change in free energy (ΔG°) is found to be negative at the studied temperatures. The negative value of ΔG° was found to increase with increase in temperature for AWR, which indicates that the sorption of BG is more spontaneous at higher temperatures. While in the case of WR the ΔG° values tended to decrease with increase in temperature, the decrease in spontaneity can be attributed to weakening of adsorption sties due to increase in temperature. The negative ΔH° values indicate that the process is exothermic in nature and negative ΔS° values indicate the decrease in the randomness of solid–liquid interface during the adsorption.

4. Conclusions

Native and activated WR was evaluated as economical sorbent for removal of BG dye from aqueous solutions. The removal for WR and AWR was found to be rapid with equilibrium been reached within 30 min and follows pseudo-second-order kinetic models. The equilibrium data tended to fit the Langmuir isotherm. The loading capacity of activated WR was found to be double than that of native WR. Thermodynamic parameters reveal that the removal of BG by WR and AWR is spontaneous and exothermic in nature. The salt ionic strength has significant effect on the adsorption of BG by WR while minimal for AWR. These observations suggest that WR can be used as a non-hazardous agro material for removal of BG dye from aqueous solutions.

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