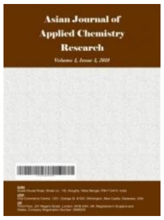

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Rice Straw and Rice Straw Ash for the Removal of Brilliant Green Dye from Wastewater

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Authors' contributions

This work was carried out in collaboration between all authors. All authors designed the study and wrote the protocol. Author GAEC managed the analyses of the study and wrote the discussion part. Authors ESR and AFA wrote the remaining parts of manuscript managed the analyses of the study and managed the literature searches. All authors reviewed and approved the final manuscript.

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ABSTRACT

Aims: The use of economical, simply obtained and green adsorbents has been employed as a perfect alternative to the expensive methods of removing dyes from aqueous solution. The capability of rice straw and rice straw ash to remove brilliant green dye by the adsorption process has been studied.
Study Design: Adsorption studies were carried out at different initial dye concentrations, contact time and adsorbent dosages.
Place and Duration of Study: Regional center for food and feed, Agricultural research center.
Methodology: Adsorption data were modeled using Langmuir, Freundlich, Temkin adsorption isotherms.
Results: Freundlich model showed the best fit with the equilibrium data for rice straw; however Langmuir model was better fitted for rice straw ash. Kinetic adsorption records were modeled using pseudo-first-order and pseudo-second-order. It was found that pseudo-second-order was best fit with the equilibrium data. Rice straw and rice straw ash were characterized by Fourier transform infrared spectroscopy (FTIR), scanning electron microscopy (SEM).
Conclusion: Rice straw as an agriculture waste by-product could be used as an alternative to commercial activated carbon as adsorbent due to its availability, removal efficiency and low cost.

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Keywords: Straw; adsorption; brilliant green; isotherm; kinetics.

1. INTRODUCTION

Rice straw is a lignocellulosic agricultural by-product composed of cellulose (37.4%), hemi-cellulose (44.9%), lignin (4.9%) and silica ash (13.1%) [1]. In rice production, it is estimated that every kilogram of grain harvested generates 1–1.5 kg of the straw [2]. The discarding of rice straw by open-field burning commonly causes severe air pollution, therefore new economical technologies for rice straw discarding and use must be exploited [3].

In Egypt, rice is one of the most abundant crops and its processing yields large amounts of rice straw as residue. About 20 % was used for other purposes such as ethanol, paper and fertilizers production as well as fodders and the remaining part is usually burnt in the fields resulting in “Black Cloud” formation [4].

Water scarcity and environmental pollutants are the most urgent problems of the 21st century. Recycling or reusing the industrial wastewater in domestic usage or irrigation is a modern trend to save water [5].

Dyes and pigments are involved in several industrial processes such as textiles, pulp and paper, food, etc. The presence of remaining dyes in the industrial wastewater effluents is undesirable. Entrance of this wastewater to a different ecosystem generates major problems to living organisms. Colored wastes in the effluents hinder sunlight transmission into the water and cause a reduction of photosynthetic activity [6].

Brilliant green (BG) is one of the commonly known cationic dyes used for various purposes, e.g.: dying silk, wool, leather, jute, cotton, a biological stain, dermatological agent, veterinary medicine, green ink manufacture, intestinal parasites, fungus textile dying and paper printing [7]. This dye is hazardous in the case of skin contact, eye contact and ingestion. It is toxic to the lungs, through inhalation. Repeated or prolonged exposure to the substance can produce target-organ damage [8]. It is thus a necessity to remove such dye from the effluents before their discharge.

Several techniques are adopted for the removal of undesired substances from wastewater. Among these techniques, the adsorption process has been widely applied for its ease of use. Also with the good choice of adsorbent, adsorption can be considered cheaper and cleaner than other treatment techniques [9].

Several studies in the literature discuss the removal of brilliant green dye from wastewater onto different natural adsorbents these include: bottom ash and deoiled soya [10], Saklıkent mud [8], *Luffa cylindrical* Sponge [11], *Psidium guajava* Leaves and *Solanum tuberosum* Peels [12], *Peganum harmala*-L Seeds [7], etc.

In the present work rice straw and its ash were studied as possible adsorbents for brilliant green dye from synthetic wastewater at different experimental conditions. The applicability of equilibrium and kinetic models was also assessed.

2. MATERIALS AND METHODS

2.1 Preparation of Adsorbent

Rice straw was obtained from a local farm in Giza governorate, EGYPT. It was thoroughly washed with water to remove dust and soluble material and dried in a hot air oven at 105ºC, then ground. A part was sieved to 250 μm to be used as it is (RS) and the other part burnt in a muffle furnace at 600ºC to ash (RSA) and stored at room temperature.

2.2 Adsorbent Characterization

FTIR spectra of rice straw (RS) and rice straw ash (RSA) were recorded using Perkin Elmer Spectrophotometer in the range from 450 to 4000 cm^{-1} with a resolution of 1cm^{-1} for each scan. Adsorbent samples were also analyzed by scanning electron microscopy (SEM) in a Jeol microscope, model JEOL JSM 6060.

2.3 Effect of Contact Time and Adsorption Kinetics

Batch experiments were done to study the effect of contact time on brilliant green dye (CI = 42040, chemical formula =C27H34N2O4S, FW= 482.62) adsorption. At the start of each batch experiment a known volume of brilliant green dye solution at the concentration of 40 mg/L was mixed with RS or RSA at a dose of 3.75 g/Lin conical flasks. The flasks were shaken at 200 rpm and monitored for different time intervals (15, 30, 60, 90, 120, 150 and 180 min) till the equilibrium was achieved. After equilibrium the solution was filtered and the remaining dye concentration was measured using a UV/vis spectrophotometer, model: Specor D250 plus; Aanlytik Jena.

A standard solution of the dye was taken and the absorbance was determined at different wavelengths to obtain a plot of absorbance versus wavelength. The wavelength corresponding to maximum absorbance (λmax) as determined from this plot was 638 nm. This wavelength was used to determine the initial and final dye concentrations during the experiments.

The adsorption capacity of BG on adsorbent and its removal percentage were calculated according to the following equations:

$$Q\ (mg/g) = (C_i - C_f)^{*}\ V/W$$

$$Removal\ \% = (C_i - C_f)/C_i \times 100$$

Where: C_f (mg/L) is the remaining concentration of dye in solution after adsorption, C_i (mg/L) is the initial dye concentration, V (L) is the volume of solution used in the experiment and W(g) is the adsorbent weight.

Two Kinetic models, namely: Lagergren's first order (eq.1) and pseudo-second-order (eq.2) were applied to fit the experimental data.

$$Ln\ (q_e - q_t) = lnq_e - k_1 t \quad (1)$$

$$t/q_t = 1/k_2 q_e^2 + 1/q_e\ t \quad (2)$$

Lagergren's first order constants are q_e is the amount of dye adsorbed onto the adsorbent at equilibrium (mg/g), q_t is the amount of dye adsorbed onto the adsorbent at any time t (mg/g), and k_1 (min^{-1}) is the rate constant of the pseudo-first-order adsorption which can be calculated from the slope of the linear plot of ($lnq_e - q_t$) against t [13]. Whereas pseudo-second-order constant are k_2 (g $mg^{-1}min^{-1}$) is the rate constant of the pseudo-second-order adsorption, q_e is the amount of dye adsorbed on the adsorbent at equilibrium (mg/g), and q_t is the amount of dye adsorbed on the adsorbent at any time, t (mg/g). k_2 (g.$mg^{-1}min^{-1}$) can be calculated from the slope and intercept of the plot of t/q_t against t [14].

2.4 Effect of Adsorbent Dose on Dye Removal

The effect of adsorbent dose on the removal % of BG dye from aqueous solution onto RS and RSA adsorbent was investigated by mixing different amounts of RS and RSA (1.25, 2.5, 3.75 and 5 g/L) with a constant concentration of BG (40 mg/L) at room temperature (25ºC) at a constant shaking of 200 rpm were tested for this study.

2.5 Effect of Dye Concentration and Equilibrium Modeling

The adsorption capability of the RS and RSA were evaluated by using brilliant green in adsorption experiments. A certain amount of adsorption material was put into a concentration of (20, 40, 60, 80, 100 mg/L) dye solution at 25ºC.

Three equilibrium models Langmuir (eq.3) [15], Freundlich (eq.4) [16] and Temkin (eq.5) [17] isotherms were used to fit the experimental data. Langmuir constants from equations are C_e (mg/L) and q_e (mg/g) are the liquid phase concentration and solid phase concentration of adsorbate at equilibrium, respectively, and q_0 (mg/g) and b_L (L/mg) are the Langmuir isotherm constants. In Freundlich equations, K_f is the Freundlich constant [mg/g $(L/g)^{1/n}$] related to the bonding energy, and n is the heterogeneity factor. b_t and a_t are isotherm constants related to the adsorption capacity of the adsorbent in Temkin model.

$$C_e/q_e = l/b_L q_0 + C_e/q_0 \quad (3)$$

$$\log q = \log K_f + 1/n \log C_e \quad (4)$$

$$q_e = a_t + b_t lnC_e \quad (5)$$

3. RESULTS AND DISCUSSION

3.1 Effect of Contact Time on Dye Removal by Rice Straw and Rice Straw Ash

In the present study contact time was investigated to determine its effect on the amount of BG adsorbed at various time intervals by a fixed amount of the adsorbent (5 g/L) at room temperature (Table 1). At the start of the experiments the removal of BG by RS and RSA increased rapidly during the first 30 min for RS and the first 60 min. for RSA. After that there was a slow increase in the removal for both adsorbents. Finally the equilibrium was reached after 150 min. for RS and 120min. for RSA and the removal of BG remained almost unchanged.

The rapid adsorption at the initial contact time can be attributed to the availability of the empty reactive site of adsorbent, while at higher time the remaining vacant surface sites are difficult to be occupied due to repulsive forces between the solute molecules on the solid and bulk phases and slow pore diffusion or saturation of adsorbent [7].Concomitantly, the contact-time between the adsorbate and the adsorbent is important in the dye-removal from the solution by the adsorption process [8].

3.2 Effect of Adsorbent Dose on Dye Removal by Rice Straw and Rice Straw Ash

In adsorption process, the adsorbent dose is a key parameter as it determines the capacity of an adsorbent for a given initial concentration of the adsorbate [18]. The results for BG removal by RS and RSA are given in Table 2. Unsurprisingly, the adsorption percentage increased with increasing the amount of adsorbent. As the adsorbent dose increased from 1.25 g/L to 5g/L for RS or RSA, the removal percentage increased from 18% to 85% (RS) or from 75% to 85% (RSA), respectively. This behavior is related to the increased number of sites available for dye adsorption on the adsorbent when increasing its dose [19]. On the other hand the adsorption capacity q (mg/g) varied considerably; the maximum capacity (8.76 mg/g) was achieved with 2.5 g RS, and 24.04 mg/g with 1.25 g RSA. The decrease in capacity per unit adsorbent with additional doses of the adsorbent is due to adsorption sites remaining unsaturated in the adsorption reaction [18].

3.3 Effect of Dye Concentration on Its Removal by Rice Straw and Rice Straw Ash

The results depicting the effect of initial BG concentration on its adsorption by RS and RSA are shown in Table 3. By increasing the initial dye concentration from 20 mg/L to 100mg/L the removal percentages decreased from 88.42 to 52.76% for RS and from 89.25 to 78.63% for RSA. These results could be attributed to the lower competition for the sorption surface sites at lower concentration. At higher concentrations, the competition for the surface active sites will be high and consequently lower sorption rates are obtained [20].

Comparing the efficiency of RS and RSA in removing BG dye from wastewater, it can be inferred from the result in (Tables 1-3) that under the same conditions RSA is slightly more efficient than RS. The maximum capacity was obtained by RSA at lower contact time (120 min.) and lower adsorbent dosage (1.25 g/L) compared to RS.

Table 1. Effect of contact time on brilliant green removal by rice straw and rice straw ash

Contact time (min.)	RS		RSA	
	Removal %	Capacity (mg/g)	Removal %	Capacity (mg/g)
15	40.734	3.258	52.231	4.178
30	57.518	4.601	54.000	4.320
60	80.318	6.425	78.671	6.293
90	82.959	6.636	84.488	6.759
120	83.837	6.706	84.803	6.784
150	84.663	6.773	84.897	6.791
180	84.655	6.772	85.018	6.801

Table 2. Effect of adsorbent dose on brilliant green removal by rice straw and rice straw ash

Adsorbent dose(g/L)	RS		RSA	
	removal %	Capacity (mg/g)	removal %	Capacity (mg/g)
1.25	18.015	5.765	75.133	24.042
2.5	54.797	8.767	83.590	13.374
3.75	59.984	6.398	83.107	8.864
5	85.018	6.801	84.655	6.772

3.4 Fourier Transform Infrared Analysis of Rice Straw and Rice Straw Ash

To get better insight into the surface functional groups present on the surface of rice straw and its ash, FTIR spectra of RS and RSA before adsorption are presented in Figs. 1 and 2, respectively. Also the spectra of RS and RSA after adsorption are presented in Figs. 3 and 4, respectively.

The spectra in Figs. 1 and 2 show bands characteristic for surface hydroxyl groups and chemisorbed water at 3427.85 and 3444.24 cm^{-1} for RS and RSA, respectively [21]. Also the spectra indicate the presence of a peak around 2920 cm^{-1} indicative for stretching of OH groups bound to methyl radicals which are common in lignin [22]. The peaks at 1071.26 and 1093.44 cm^{-1} in RS and RSA spectra, respectively, are due to Si-O-Si bond [23]. The RS spectrum shows a sharp peak around 1635$cm^{-1}$for -CO and –C-OH groups stretching from aldehydes and ketones [24]. On the other hand, the spectrum of RSA gives a strong peak at 791.63 cm^{-1} for Si-H group while the spectrum of RS shows a peak at 463.796 $cm^{-1}$for bond of Si-O-Si bending [25].

After the adsorption experiments, the FTIR spectra of RS and RSA loaded with BG were recorded as shown in Figs 3 and 4. The peaks present in the adsorbents spectra before their uses were shifted to other wave numbers, split or disappeared after adsorption. The FTIR spectrum for RS loaded with BG shows broadening with little shift of the band at 3427.85 cm^{-1}. Also the bands at around 1635 and

Table 3. Effect of dye concentration on its removal by rice straw and rice straw ash

Dye concentration (mg/L)	RS		RSA	
	Removal %	Capacity (mg/g)	Removal %	Capacity (mg/g)
20	88.4203	3.4100	89.252	3.536
40	84.6053	6.2616	85.513	6.768
60	73.4667	10.592	82.406	8.816
80	59.5608	13.117	81.985	12.551
100	52.7674	15.727	78.635	10.553

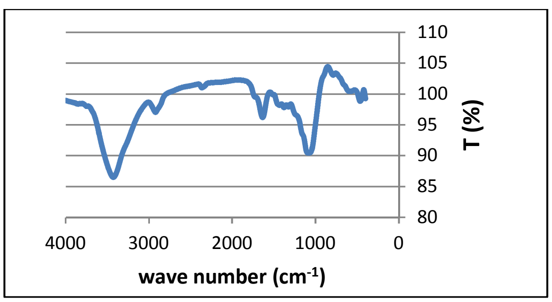

Fig. 1. FTIR spectrum of rice straw (RS) before adsorption

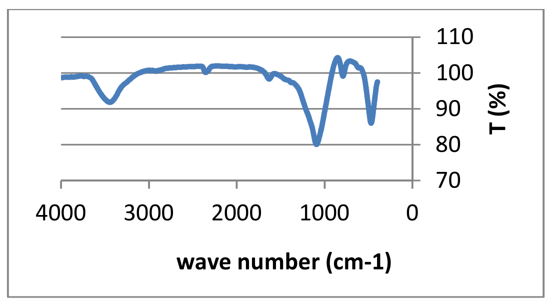

Fig. 2. FTIR spectrum of rice straw ash (RSA) before adsorption

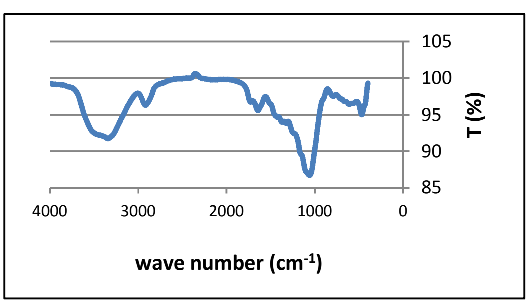

Fig. 3. FTIR spectrum of rice straw (RS) after adsorption

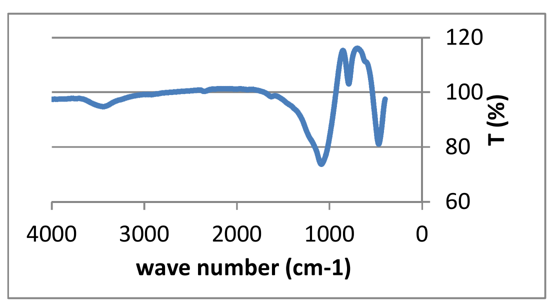

Fig. 4. FTIR spectrum of rice straw ash (RSA) after adsorption

1071 cm^{-1} were shifted after adsorption. Similarly, the spectrum of RSA after adsorption showed disappearance of the bands at 3444, 2920, 1632 cm^{-1} and shifting of the peaks at 1093, 791 and 471 cm^{-1}. These results suggest the involvement of several adsorbents' functional groups in the removal of BG dye.

3.5 Scanning Electron Microscopic (SEM) Analysis of Adsorbents

The SEM micrograph of rice straw Fig. 5 (a) shows irregular plates and cracks on the surface while the SEM of rice straw ash Fig. 5 (b) shows that it has a porous structure. This surface morphology of the RS and RSA gives an idea about their possible adsorption capabilities.

3.6 Equilibrium Modeling for Adsorption of Brilliant Green by Rice Straw and Rice Straw Ash

The equilibrium modeling of adsorption plays an essential function in the determination of the maximum capacity of adsorption. The adsorption equilibrium data of BG on rice straw and rice straw ash were evaluated by Langmuir, Freundlich and Temkin models.

Table 4 gives the regression coefficients and the calculated parameters for the Langmuir, Freundlich and Temkin models.

Since the value of the correlation coefficient (R^2) nearer to 1indicates that the respective equation better fits the experimental data, as seen from Table 4, the adsorption data of BG by RSA were best fitted by the Langmuir model which suggests that adsorption takes place by monolayer adsorption on a homogeneous surface. The Langmuir maximum capacity (q_0) was found to be 11.628mg/g and the Langmuir constant b_L (L/mg) was found to be 0.2723. The essential characteristics of Langmuir isotherm can be described by a separation factor (R_L), which is defined as ($R_L = 1/(1+bC_i)$), where C_i is the initial concentration of dye (mg/L) and b_L is Langmuir constant which indicates the nature of adsorption. The separation factor R_L indicates the isotherm shape and whether the adsorption is favorable or not [R_L>1 (Unfavorable), R_L =1 (linear), 0 < R_L<1 (favorable), R_L =0 (irreversible)] [26]. In the present work the R_L values of the studied concentration range were found to be (0.035 – 0.155) indicating favorable adsorption of BG on RSA.

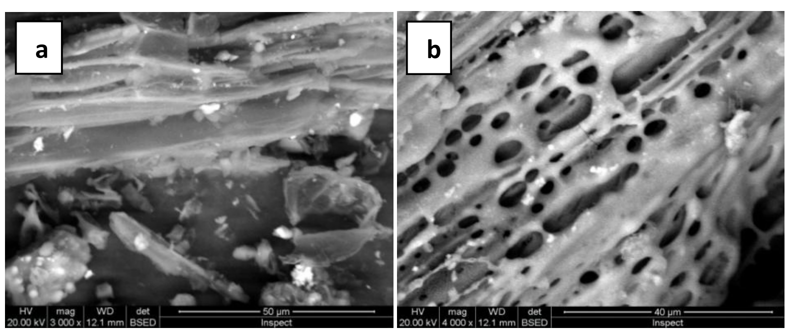

Fig. 5. Scanning electron micrograph of rice straw (a) in 3000x resolution and rice straw ash (b) in 4000 X resolution

Table 4.Comparison of various isotherm equations for the adsorption of brilliant green by rice straw and rice straw ash

Isotherm models	Parameters	Rice straw	Rice straw ash
Langmuir	**q_0(mg/g)**	30.675	11.628
	b_L	0.052	0.272
	R^2	0.783	0.975
Freundlich	**K_f**	0.893	0.316
	n	1.243	2.591
	R^2	0.958	0.794
Temkin	**a_t**	3.381	2.093
	b_t	6.042	2.618
	R^2	0.829	0.740

q_0 and b_L: Langmuir isotherm constants;R^2:Correlation coefficient; K_f: Freundlich constant; n: The heterogeneity factor; a_t and b_t: Isotherm constants

Table 5. Comparison of various kinetic equations for the adsorption of brilliant green by rice straw and rice straw ash

kinetic models	Parameters	Rice straw	Rice straw ash
Lagergren's pseudo-first order	k_1	0.048	0.039
	q_e	6.611	5.766
	R^2	0.961	0.971
pseudo-second-order	k_2	0.009	0.008
	q_e	7.462	7.518
	R^2	0.995	0.996

k_1: Rate constant of the pseudo-first-order adsorption; q_e: Equilibrium capacity; R^2: Correlation coefficient; k_2: Rate constant of the pseudo-second-order adsorption

Whereas, the adsorption of BG by RS was better described by the Freundlich model suggesting heterogeneous sorption. The values of n and K_F were 1.24 and 0.8938 respectively. The value of 1/n less than 1 shows the favorable nature of adsorption of BG on RS [27].

The Temkin model did not show any good fit for the adsorption data of BG by both RS and RSA.

3.7 Kinetics Study of Brilliant Green Adsorption on Rice Straw and Rice Straw Ash

To investigate the possible mechanism of adsorption, pseudo-first-order and pseudo-second-order adsorption models were tested to fit the experimental data. The kinetic results for the adsorption of BG by RS and RSA are given in table 5. The results showed that the adsorption processes of BG by rice straw and its ash follow the pseudo-second order kinetic model with R^2>0.99. The pseudo-second-order model is based on the assumption that the adsorption process is chemisorption [19,28]. These results are in accordance with previously reported data for adsorption of dyes on various other non-conventional adsorbents [29].

4. CONCLUSION

Rice straw and rice straw ash were applied as adsorbents for the removal of brilliant green dye from aqueous solution. The two adsorbents showed a good adsorption capacity to remove the dye. The removal efficiencies were affected by the contact time, initial dye concentration and adsorbent dose. The adsorption of brilliant green was best fitted by the Langmuir model for rice straw ash and Freundlich model in case of rice straw. The pseudo-second-order kinetic model fitted very well the adsorption behavior of brilliant green dye. This agriculture waste by-product could be used as an alternative to commercial activated carbon as adsorbent due to its availability, removal efficiency and low cost.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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