Utilization of rice hulls for adsorption of methylene blue dyes from aqueous solution

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The removal of dyes from colored effluents, particularly from textile industries, is one of the major environmental concerns these days. Current methods for removing dyes from wastewaters are costly and cannot effectively be used to treat wide range of such wastewaters. This work describes the use of grounded rice hull as adsorbent material. Aqueous solutions of various concentrations (5-25 mg L^{-1}) were shaken with certain amount of adsorbents to determine the adsorption capacity both treated and untreated rice hulls were used for methylene blue adsorption. The effects of adsorbents dose, initial pH, initial dye concentration and contact time on dye removal have been studied. Maximum dye was sequestered from the solution within 60-90 min after the beginning of every experiment. The adsorption capacity increased from (72 to 94 %) with increasing the pH from (3 to 10). Pretreatment of rice hulls with citric acid did not reveal any beneficial effect. Rice hulls were more effective compared to commercial used adsorbents. The results showed that ground rice hulls can be considered as potential adsorbents for methylene blue removal from dilute aqueous solutions.

التخلص من الوان الصبغات من المحاليل وخاصة اثناء صناعة الملابس يعتبرواحد من اهم الاتجاهات البيئية اليوم. الطرق المستخدمة لازالة الصبغات اثناء معالجة المياه تعتبر مكلفة ولايمكن استخامها لمعالجة كميات من هذه المياه لذا كان الاتجاه في هذا البحث لاستخدام قشرة الارز كمادة مازة وذلك بتحضير محاليل من صبغة الميثيلين الزرقاء بتركيزات تتراوح بين ٥-٢٥ مللي جرام/لتر مع استخدام كميات مختلفة من قشرة الارز وذلك لدراسة قدرة القشرة علي الامتصاص قبل وبعد المعالجة. وقد تم دراسة تاثير كمية المادة المازة، الاس الهيدروجيني، تركيز الصبغة وزمن الاستبقاء ووجد ان اعلي نسبة من الصبغة تم ازالتها في ٦٠-٩٠ دقيقة من بداية التجربة وتزيد قدرة الامتصاص من ٧٢ الي ٩٤ % وذلك بزيادة الاس الهيدروجيني من ٣-١٠. معالجة قشرة الارز باستخدام حامض الستريك لايؤثر علي كفائته في ازالة الصبغة. بمقارنة قشرة الارز ببعض المواد المازة المستخدمة تجاريا اوضحت النتائج ان للقشرة قدرة امتصاص اعلي لذا ينصح باستخدامه في ازالة صبغة الميثيلين الزرقاء وذلك لانه من المخلفات الزراعية عديمة الفائدة والتي تتحلل طبيعيا فلا يسبب مشاكل بيئية.

Keywords: Bioadsorbent, Rice Hulls, Methylene blue, Agriculture waste

1. Introduction

Lignocellulosic agricultural waste materials such as rice hulls, sugarcane bagasse, and wheat straw are regarded as abundant, inexpensive, and readily available natural resources for chemical and paper industries [1, 2]. In 1999, the world's industry is utilizing less than 10 % of biomass from plantations [3]. In Egypt, approximately 16 million tons of Agricultural Residues (AR) are currently disposed every year, the annual amount of rice hulls is about 3.6 million tons/year [4] and many types of abundant AR especially from tropical plantations are waiting for effective utilization. AR are the most abundant renewable organic resource on the earth. AR have been utilized as renewable resources of energy and production of a diversity of chemicals, including ethanol [4], activated carbon [5], ion exchangers [6]. Many others application for AR are in the process of being developed. Annual world rice production is about 577 million metric tons [7]. The rice hulls which are discharged in large quantities as rice processing facilities or country elevators in rice cropping regions are utilized as farmyard compost, mulch and drainage materials, but the recycling rate of the rice hulls is about 10.0% [8]. However, many countries have imposed new regulations to restrict field burning of rice hulls in response to restrictions on carbon emission due to global warming. This has helped simulate interest in the utilization of rice waste as a renewable natural resource. Various techniques have been employed for the removal of dyes from wastewaters [9-17]. Conventional physical and chemical methods are either costly, e.g. Activated carbon, or produce concentrated sludge, e.g. Fenton's reagent, or may not be capable of treating large volumes of effluent without the risk of clogging, e.g. membrane filtration [18]. Natural materials that are available in large quantities, or certain waste product from industrial or agricultural processes, may have potential as inexpensive adsorbents. Rice hulls, an agricultural waste, have been reported as a good adsorbent of heavy metals [19, 20]. The focus of the research is to evaluate the adsorption capacity of treated and untreated rice hulls for the adsorption of methylene blue dye from aqueous solutions. Methylene blue is the most commonly used material for dying cotton, wood and silk. Methylene blue was chosen because of its known strong adsorption onto solids and it serve as a model compound for removing organic contaminants and colored bodies.

2. Experimental work

2.1. Materials

Rice hulls were used as precursor material of adsorbents. Rice hulls were obtained from local rice mills (Behera city - Egypt). The methylene blue dye (molecular formula: $C_{16}H_{18}N_3ClS$, λ_{max}= 655nm) used in this study has been obtained from Nice chemicals Pvt.Ltd.

Commercial adsorbents were also used for comparison table 1.

2.2. Preparation of adsorbent

Rice hulls were received from local rice mills grounded and then start to prepare samples.

2.2.1. Untreated rice hulls

Rice hulls were washed four times with tap water, and then dried at 105 ^{O}C for 2h. The rice hulls were sieved to 250-500 µm, and used for the study.

Table 1
Types of commercial adsorbents used

Adsorbent type	Source	Composition	Appearance	Color	Specifications
1-Adsorb-it Filtration Fabric	Eco-Tec, Inc, U.S.A.	A geo- textile quality non-woven filtration fabric manufactured from 100% recycled select fibers from the textile industry.	Thin sheet, no odor	White gray	thickness 3.73 mm
2-Adsorb-it/AM Antimicrobial Filtration Fabric	Eco-Tec, Inc, U.S.A.	Fabric of Adsorb-it treated with a surface bonded non-leaching antimicrobial.	Have two surfaces one is more rough than the other one, no odor, thin sheet	Dark gray	thickness 3.8 mm
3-Oil-Only sorbent	ARCUS Absorbents Inc, Toronto, Ontario Canada	It consists of a hydrophobic polypropylene product.	light weight, flame resistant, non-toxic	White color	Available pad form with size 17″×19″×0.69″.

2.2.2. Treated rice hulls

Rice hulls were washed four times with tap water, and then dried at 105 ^{O}C for 2h.

Soaking it for 2h in citric acid 0.6M at room temperature was applied to activate the rice hulls which were then sieved to 250-500 μm. The acid- hulls slurry was dried over night at 50°C then the hulls were boiled at 100 °C for one hour and then washed with distilled water (200 ml/g of hulls) to remove any excess of citric acid followed by overnight drying at 105 °C.

2.3. Preparation of dye solution

A stock solution of the dye was prepared by dissolving 1.0 g of dye in 1000 ml distilled water to make a stock solution of 1000 mg L^{-1}. The experimental solution was prepared by diluting definite volume of the stock solution to get the desired concentration. For absorbance measurements a spectrometer UV-VIS double beam PC scanning (Lambomed.INC) was employed, the rice hulls was gravity separated before sampling .The maximum wavelength λ_{max} for the methylene blue was measured at 655 nm. Concentrations during experimental work were determined from a standard calibration curve.

2.4. Adsorption studies

Equilibrium adsorption isotherms using raw and treated rice hulls with different amount (5, 10, 15 g/L) were determined at agitation rate of 150 rpm, at room temperature and with different concentrations of dye (5 - 25 mg L^{-1}). Adsorbent dosage (based on dry weight of rice hulls) was placed in a set of 100 mL beaker and different initial dye concentrations were added to each beaker. The contents were shaken for 2 hours the solution was analyzed for dye contents. The adsorption behaviors of the samples were studied by evaluating the percentage removal efficiency of methylene blue, calculated as

$$\text{Removal efficiency} = [(C_0 - C)\ C_0] \times 100$$

Where C_0 is the initial concentration of methylene blue, C is the solution concentration after adsorption at any time. The effect of adsorption time on the dye removal at various predetermined intervals from (10-120 min) was monitored by shaking the reaction mixture and analyzed for the dye content at the end of each contact time.

2.5. Equilibrium studies

The amount of adsorption for untreated and treated rice hulls, q_e (mg/g), was calculated by:-

$$q_e = (C_0 - C_e)\ V / W$$

where C_0 and C_e (mg/L) are the liquid-phase concentrations of dye at initial and equilibrium, respectively. V is the volume of the solution (L) and W is the mass of dry sorbent used (g).

2.6. Scanning electron micrograph and FTIR studies

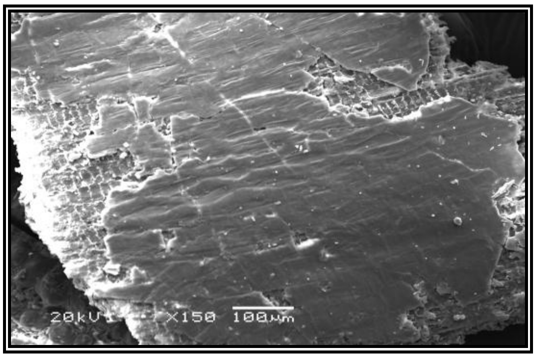
SEM ("JEOL JSM 6360LA"- Japan) was used to study the morphological features and surface characteristics of the adsorbent materials. (Nelly. et al., Gupta.S.et al.) In this study, SEM is used to study and assess morphological changes in the rice hulls surfaces treated and untreated following adsorption of the dyes molecules also the fibers were also analyzed with fourier transform for infra red spectrophotometer (FT IR -8400S Shimadzu) in order to identify the function groups in the fiber structure.

3. Result and discussion

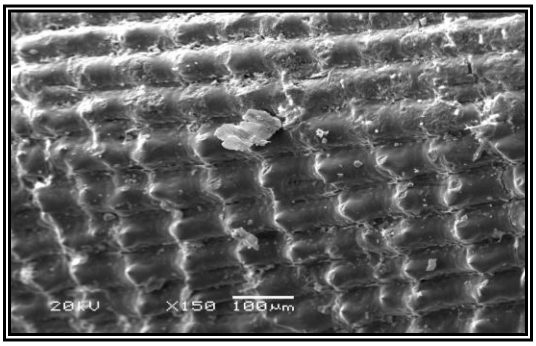
3.1. Morphology study

3.1.1. Morphology of unloaded rice hulls

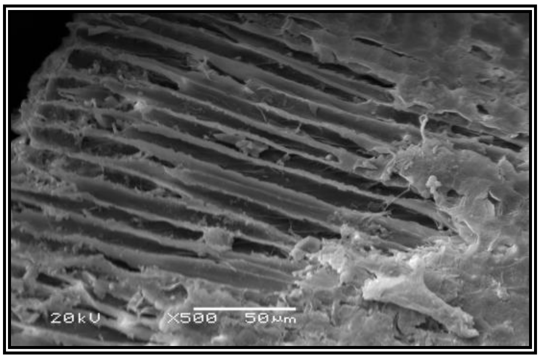
Figs. 1-a, b and 2-a, b of untreated and treated adsorbents indicates its morphological features. The SEM photographs for the rice hulls showed cellular structure and it retain cellular structure of the parent material after treating with citric acid but it is more attacked. Especially the intercellular wall.



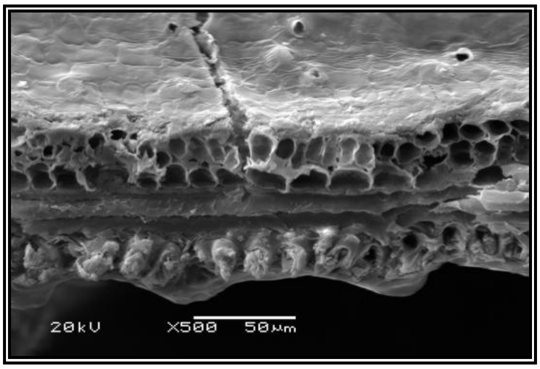
(1-a)



(1-b)



(2-a)



(2-b)

Fig. 1, 2. Scanning electronic micrographs of untreated and treated rice hulls.

3.1.2. Morphology of dye loaded adsorbent

The morphology of the loaded adsorbent showed blocking of the tubular tube in the structure of untreated and treated rice husk reveal entrapping the dye molecule, the above observation was further confirmed well with the adsorption studies. Figs. 3-a, b.

3.2. FTIR study

The FTIR spectrum fig. 4 shows band at 3462 cm^{-1} due to –OH stretching. The band at 2941 cm^{-1} corresponds to C-H asymmetric stretching of $–CH_2–$ groups. The band at 1639 cm^{-1} is attributed to H-O-H bending. it can be seen from the FTIR spectrum of treated rice hulls that the intensity of the band at 3462 cm^{-1} present in rice hulls due to –OH stretching has considerably increased after treatment with citric acid, two new bands appeared at 1251 cm^{-1} and 243 cm^{-1} which are of C-O stretching and provide evidence that the citric acid has been estrified with the –OH group of rice hulls to form cell –OCOC moiety [21].

3.3. Effect of adsorbent mass

The effect of adsorbent amount on the uptake of the dye was measured for dye concentration 10 mg L^{-1}, different shaking time (10-240 min) and different quantity of rice hulls (5, 10, and 15g) at pH 7 and room temperature (26 ±1 °C). Fig. 5-a, b. In general, it was found that by increasing the amount of the adsorbent the adsorption rate increased. Maximum dye removal was achieved within 60-90 minutes after which a decrease in methylene blue concentration was negligible. Increase in dye removal percentage with adsorbent dose can be attributed to increased adsorbent surface area and availability of more adsorption sites. At the beginning of the process the rate of dye removal by the rice hulls was fast during the first 30 min and then decrease gradually.

3.4. Comparison of treated and untreated rice hulls

The effect of treating the rice hulls was studied at fixed adsorbent dose 10 g hulls, temperature (26 ±1 °C), with initial concentration of methylene blue 10 mg L^{-1} and pH neutral for different time intervals. It is evident from fig. 6 that the adsorption of the dye with treated hulls was higher than the untreated and equilibrium was established in about 90 min.

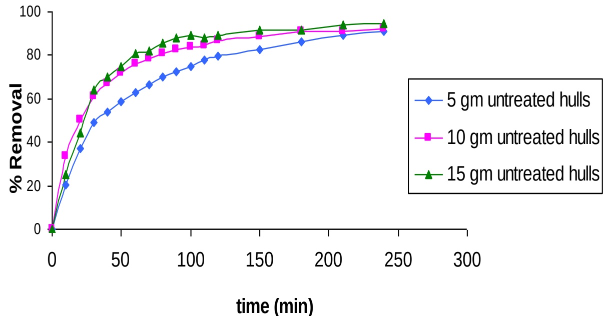
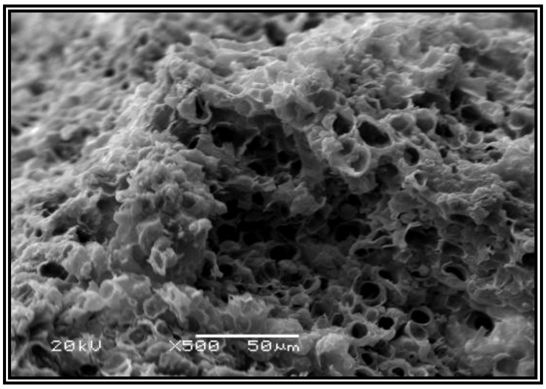
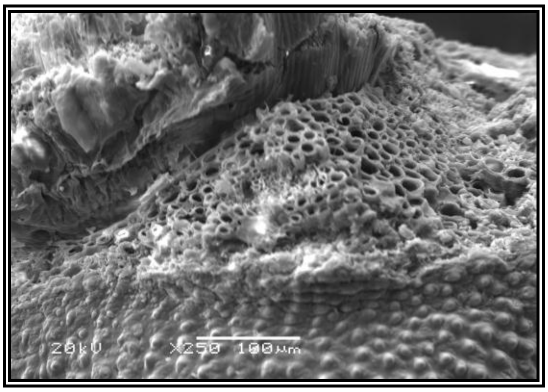


Fig. 5-a. Effect of amount of untreated rice hulls on the removal of dye.



(3-a)



(3-b)

Fig. 3-a, b. Scanning electronic micrographs of
a) loaded untreated rice hulls with dye
b) loaded treated rice hulls with dye.

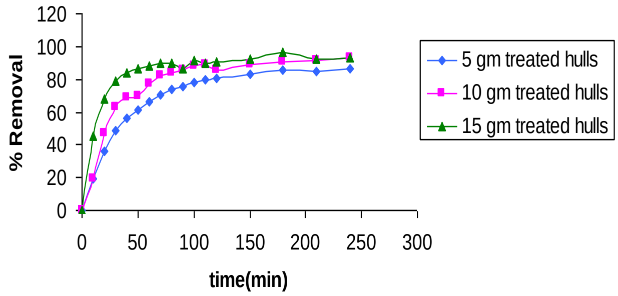


Fig. 5-b. Effect of amount of treated rice hulls on dye removal.

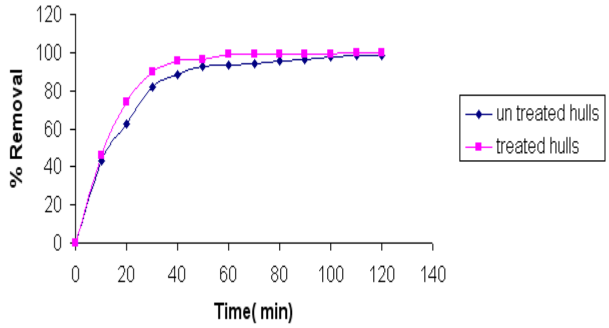


Fig. 6. Effect of chemical treatment of rice hulls on methylene blue adsorption.

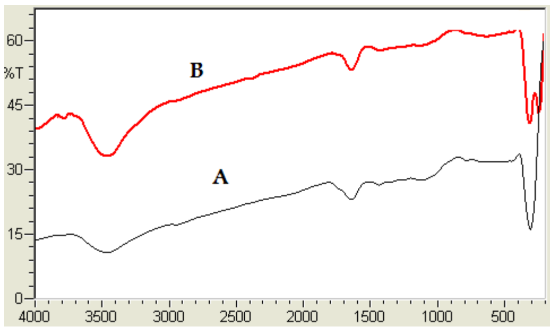


Fig. 4. FTIR spectra of rice hulls (A) Untreated, (B) Treated.

3.5. Effect of initial dye concentration

The influence of the initial concentration of methylene blue (5-25 mg L^{-1}) on the adsorption rate using untreated and treated rice hulls was studied. The experiments were carried out at fixed adsorbent dose (10 g), at room temperature (26 ±1 °C), neutral pH (7.0) for 30 min.

Dye removed by treated and untreated rice hulls were about 90% for low concentration (5g/L) after 30 min. The removal efficiency of the two adsorbents decreased with increasing initial dye concentration.

Table 2 shows that the percent adsorption decreased with increase in initial dye concentration, but the actual amount of dye adsorbed per unit mass of adsorbent increased with increase in dye concentration in the test solution for different concentrations (5-25 mg/L) at equilibrium (q_{max}) the same time (min).

The unit adsorption for untreated rice hulls increased from 0.41 mg/g to 1.57 mg/g as the methylene blue concentration in the test solution was increased from 5 to 25 mg L^{-1}. Similarly, unit adsorption for treated rice hulls increased from 0.45 mg/g to 1.78 mg/g as the methylene blue concentration in the test solution increased from 5 to 25 mg L^{-1}. Maximum dye was sequestered from the solution within 15 min (30 or 90 as previously) after the beginning for every experiment. Thereafter, the decrease of methylene blue concentration was negligible.

3.6. Effect of pH

The pH value of the solution is an important parameter for the adsorption processes, and the initial pH value of the solution has significant influence compared to the final pH [22] .To study the effect of pH on methylene blue adsorption, the experiments were carried out at 10 mg L^{-1} initial dye concentration with 10 g/L adsorbent dosage at (26 ±1 °C). In general, initial pH value may enhance or depress the uptake. This is attributed to the charge of the adsorbent surface with the change in pH value. Fig. 7 shows the relationship between the pH value and the removal of methylene blue. It can see from the figure that as the solution pH increases, the adsorption capacity increases. Increasing solution pH increases the number of hydroxyl groups thus, increases the number of negatively charge sites and enlarges the attraction between dye and adsorbent surface, [23]. Generally, the net positive charge decreases with increasing pH value lead to the decrease in the repulsion between the adsorbent surface and the dye thus, improving the adsorption capacity.

3.7. Isotherm analysis

The purpose of the adsorption isotherms is to relate the adsorbent concentration in the bulk solution and the adsorbed amount at the interface [24] .The equilibrium isotherms in this study have been described in terms of Freundlich isotherms.

The Freundlich isotherm [25] is an empirical equation assuming that the adsorption process takes place on heterogeneous surfaces and adsorption capacity is related to the concentration of methylene blue dye at equilibrium. A linear form of the Freundlich equation is generally expressed as follows:

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

Table 2
Effect of methylene blue concentration on dye adsorption

Initial dye concentration (mg/L)	Percent dye removal with time(min)					
	10	20	30	40	50	60
Untreated rice hulls						
5	65.46	80.22	82.86	89.23	90.2	98.84
10	42. 9	62.18	82.1	88.23	92.9	98.59
15	41.97	53.57	67.63	74.84	80.19	86.38
20	41.16	58.82	70.185	75.23	79.69	85.37
25	38.67	56.96	62.98	66.23	66.99	72
Treated rice hulls						
5	81.1	85.2	89.98	96.35	98.8	98.97
10	46.2	73.9	89.85	95.7	96.25	97.8
15	36.63	65.89	78.57	80.01	86.32	90.65
20	29.04	48.46	68.92	73.43	78.25	80.22
25	32.02	54.51	71.28	75.60	78.06	79.34

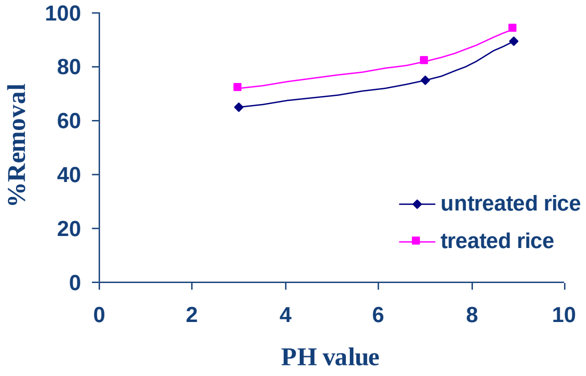
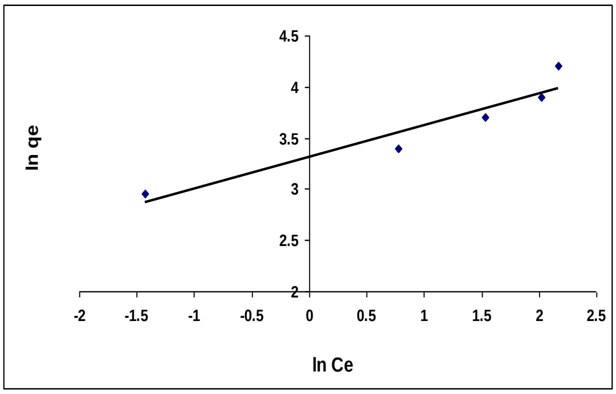


Fig. 7. Effect of pH on methylene blue adsorption.

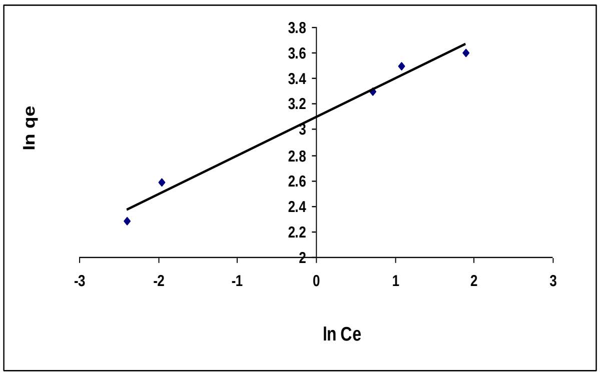
where K_F (mg/g (L/mg)$^{1/n}$) is roughly an indicator of the adsorption capacity and $1/n$ is the adsorption intensity. The magnitude of the exponent, $1/n$, gives an indication of the favorability of adsorption. Values of $n > 1$ represent favorable adsorption condition [26]. The plot of ln q_e versus ln C_e fig. 8-9 is employed to evaluate the intercept K_F and the slope $1/n$. The values of K_F, n and the linear regression correlation (R^2) for Freundlich are given in table 3.

3.8. Comparison between the raw, treated rice hulls and commercial adsorbents

The adsorption of methylene blue onto three different commercial adsorbents , treated and untreated rice hulls was studied at fixed adsorbent quantity (10 g), temperature (26 ±1 °C) and neutral pH for a 120 min period. It is evident from fig. 10 that the adsorption capacity of the treated and untreated rice hulls is higher compared to commercial adsorbents except for dye adsorbent which revealed higher percentage removal during the first 60 minutes and following this, the adsorption of rice hulls were almost equal, which confirm that this waste can be used as an adsorbent.



(a)



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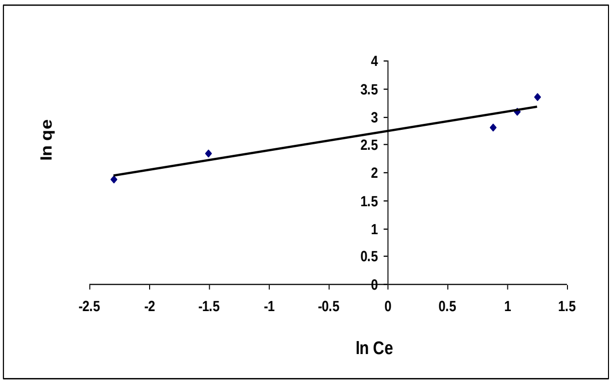
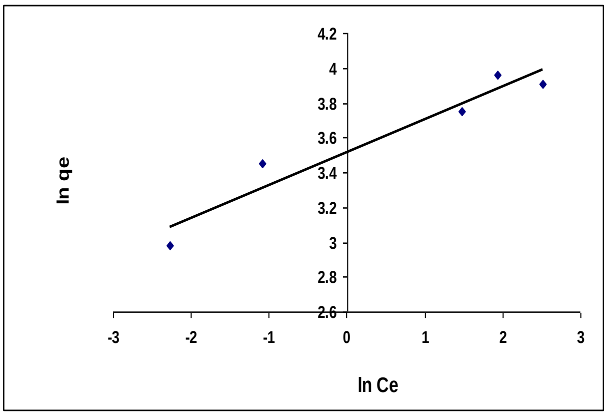
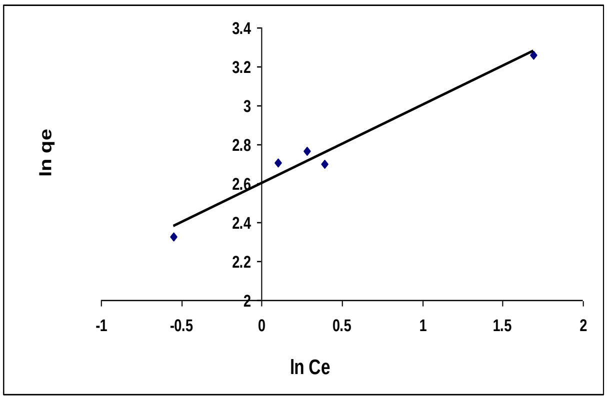


(c)

Fig. 8. Freundlich isotherm of methylene blue adsorption onto untreated rice hulls (a) 5 gm, (b) 10gm, (c) 15 gm.



(a)



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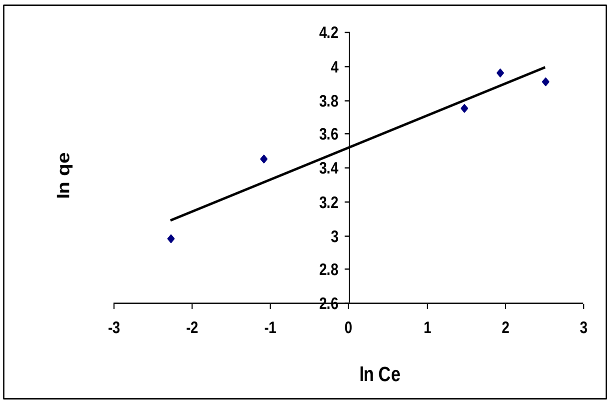

(c)

Fig. 9. Freundlich isotherm of methylene blue adsorption onto treated rice hulls (a) 5gm, (b) 10gm, (c) 15 gm.

Table 3
Isotherm parameters for removal of methylene blue by untreated and treated rice hulls

Isotherm	Parameters	
	Untreated rice hulls	Treated rice hulls
5 gm		
n	3.2	5.32
K_F	27.54	33.56
R^2	0.9075	0.9306
10 gm		
n	3.32	4.67
K_F	22.1	15.5
R^2	0.9803	0.979
15 gm		
n	2.9	2.49
K_F	15.44	13.5
R^2	0.9167	0.9677

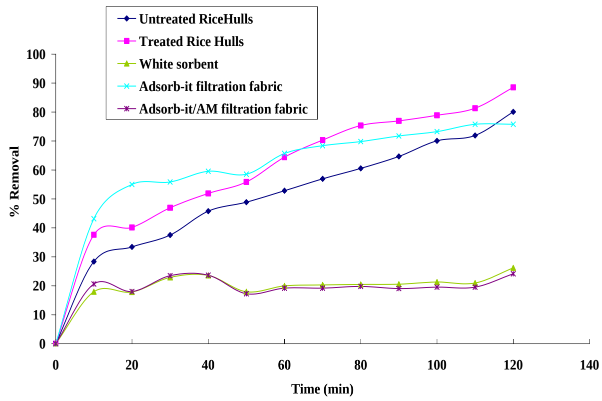

Fig. 10. Comparison between commercial sorbents and rice hulls residue treated and untreated.

3.9. Technology importance, separation technique and disposal

Natural fibers have little resistance towards environmental influences and show an intrinsic variability of their properties [27]. Rice hulls was chosen to be applied as adsorbent material due to its fibrous structure , insolubility in water, chemical stability, high mechanical strength and its local availability at almost no cost. The advantage in the application of this waste is that there is no need to regenerate them because of their low production costs [28]. The concentration of dye was measured using spectrophotometer UV-VIS double beam by gravity separation. The rice hulls which are discharged after dye removal can be used as insulating material (walls, floors or roofs) and for manufacture of particle boards.

4. Conclusions

The removal of methylene blue from wastewater using grounded rice hulls has been investigated under different experimental conditions in batch mode. The adsorption of dye was dependent on adsorbent dose and methylene blue concentration in wastewater. Initial pH of solution affected the adsorption of this dye. The higher pH for the removal of methylene blue from aqueous solution under the experimental conditions used in this work was 10. Maximum dye removal was observed within 30 min from the beginning of each experiment. No pretreatment is necessary as sufficient dye removal is achieved by the untreated material. The present investigation showed that rice hulls can be effectively used as adsorbent comparable with the three commercial adsorbents used. Rice hulls are economically cheap and so regeneration is not necessary. The data may be useful for designing and fabricating an economically cheap treatment process for the removal of methylene blue from dilute industrial waste waters.

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