RESEARCH ARTICLE

Effectiveness of Quercus Branti Activated Carbon in Removal of Methylene Blue from Aqueous Solutions

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A-B-S-T-R-A-C-T

Background & Aims of the Study: Dyes are one of the most contaminants in textile industrial wastewater that they are often carcinogenic, mutagenic and non-degradable. Therefore, with regard to environmental aspects, their removal from effluents is very essential. The purpose of this study was the perception of adsorption process and promotion of an economic technology for colored wastewater treatment. Therefore, activated carbon from Oak fruit bark was used as an effective and economic adsorbent.

Materials & Methods: This study was performed at laboratory scale and batch system. At present study, the adsorbent surface properties was evaluated by use of the (FT-IR) test and scanning electronic microscope (SEM). Also, effect of various operating parameters such as pH, contact time, adsorbent dose, initial dye concentration and temperature on dye removal from synthetic wastewater were studied.

Results: In this study, maximum removal efficiency of methylene blue were achieved at optimal pH=6, reaction time 180 minutes, and adsorbent dose 2 gl[-1]. Methylene blue removal efficiency with initial concentration of 100 mg.l[-1] was 91.08%.

Conclusions: According to results, it was cleared that : Quercus branti activated carbon can be used as an effective and economic adsorbent in waste water treatment processes.

Background

Currently there are about 10 thousand different synthetic dyes and more than 10×10^5 tons of color produced in around the world annually (1). Because of existing many dyes with high COD, low biodegradation, and their high productions, demands and consumptions in industries such as textile, rubber, plastics, pharmaceutical, paper, leather, food, cosmetic and paint, treatment and disposal of dye effluents have been an environmental serious problem (2-4). According to their application in industry, dyes can be classified into several different categories that include acidic, basic, disperse and direct (5). Many of these dyes due to their complex structure are non-

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biodegredable, toxic, carcinogenic and mutagenic (6).

Discharge dyes wastewater from the textile industrial into receptive water resources, in addition to creating an unpleasant scene and popular protest, is led to sunlight penetrating reduction into the deep layers of the receptive water; such as human visual disorder, and photosynthetic performance in aquatic plants, the occurrence of the phenomenon eutrophication (growth rapid and excessive algae), increasing wastewaters hardness and turbidity, death and destruction of aquatic organisms in surface waters and human gradual poisoning (7). Dyes wastewater treatment, due to existence of persistent and non-biodegradability of aromatic molecular compounds in the paint, is difficult and complex (8-10). Used methods for dye removal from industrial wastewaters are mostly biological, chemical and physical (11-13). Because of low biodegradable, the low ratio of BOD to COD (0.1), inhibitor effect on some bacteria and rapid change in color quality and quantity, biological methods almost could not able to remove high concentration of textile wastewater (14). Although chemical treatment by UV, H_2O_2, O_3, Photocatalyst and Fenton are very fast, they are complex, expensive methods with production of by-products (15).

Other methods, such as the precipitation, ion exchange, membrane filtration, electrochemical are used for dye removal from wastewater, but most of these methods are costly and led to production of sludge and dangerous by-products (16,17). Adsorption process is one of the physical methods (15). Recently rapid adsorption of soluble dyes from effluents on natural materials has been very remarkable. Adsorption process has more advantages than other treatment processes, including high potential in removal of tiny molecule materials (color and organic matter), low sensitivity to the fluctuations of flow, not the impact of toxic chemicals on the process, low used land (half or a quarter of a biological process), high flexibility in design and preformation of process, low initial cost, simple design, comfortable performance and insensitivity to toxic substances (18-21).

Because of high efficiency, activated carbon is one of the most important adsorbents used for the removal of dyes from aqueous solutions. Because purchase and regeneration cost of active carbon is high, its use in large scale is not economical (22-24). Therefore, discovering economic different sorbents for dye removal from aqueous solutions has been increased now. Some these sorbents are biological solids (25), wheat bran (26), sugar cane waste, corn flakes, rice bran, banana peel, the coconut bark (20,21,27-29).

Aims of the study:

According to previous studies, so far fruit Quercus branti tree has not used for any pollution removal. For this reason, we used active carbon of modified skin of the fruit Quercus branti tree, as a suitable and available absorbent for methylene blue removal from aqueous solutions under different parameters such as pH, temperature, initial dye concentration, adsorbent dose, reaction time.

Materials & Methods

Absorbent Preparation

Fruit Quercus branti was collected from forest locations in Lorestan, Iran. For preparation of active carbon, in the first oak fruit was washed completely by tap water and distillation water and then dried in 105 ^{0}C temperature for 60 min. After drying, it was divided into the same sized by standard mesh. Diameter range of produced active carbon was from 2 to 3mm. Frequently after sizing particles, produced active carbon heated in a quartz furnace under temperatures of 450, and 750 ^{0}C at time of 120 and 30 minutes respectively and N_2 gas was used for vacuum station in furnace. Produced particles were washed by distillation water and dried in oven for 60 minutes in 105 ^{0}C temperature, and then

for next consumption, they were kept in a desiccator .To examine the surface morphology of the adsorbent, the adsorbent before adsorption process, scan electronic microscopic (SEM) model "Jeol MODEL Jsm-T330" was used. In addition, the adsorbent surface characteristics were analyzed by Fourier transform infrared (FT-IR) "perkin Elmer of spectrum one model'' in limitation of 450-4000/1cm.

Adsorption experiments
In this study was used cationic dye of methylene blue (MB) (319.85 g.l^{-1}) for preparation of stock solution with concentration of 1000 mg.l^{-1} and required dye initial concentration made from this stock by dilution with distillation water. MB and others materials were purchased from Merck Co in German. Chemical properties and structure of this dye have shown in figure and table 1.

Based on batch reactor method, all of the adsorption experiments of methylene blue were performed in a 500 ml flask containing 250ml of dye sample. In this study, methylene blue removal efficiency with initial concentration of 100 mg.l^{-} was studied under parameters such as pH, adsorbent dose, reaction time, dye concentration, and temperature. Solution pH was adjusted by chloric acid and sodium hydroxide 0.1N and measured by pH meter mode HACH-HQ-USA. After mixing process by mixer with 140 rpm in ambient temperature, samples stayed in static situation for 30 min to be precipitated, and then analyzed by UV-VIS spectrophotometer "DR5000-UV-visible Model" with maximum long wave of 663 nm. Absorption kinetics of methylene blue removal were analyzed according to different isotherms. Also adsorption efficiency and adsorption capacity were detected according to following equations (Eq.1,2).

$$q_e = C_i - C_e VM \qquad (1)$$
$$\text{Adsorption (\%)} = C_i - C_e C_i \qquad (2)$$
Where:

C_i and C_e are initial concentration and dye balance after adsorption (mg.l^{-1}) respectively, and V the volume of solution (L), (M) the mass of adsorbent (g), q_e the amount of dye adsorbed at equilibrium (mg.l^{-1}).

Table 1) Physical characteristic of methylene blue

Methylene blue	The scientific name
C$_{16}$ H$_{18}$ N$_3$ CLS	Chemical formula
19.85 gl^{-}	Molecular weight
663nm	The wavelength of maximum absorption (nm)
Blue	Color
Cationic	Color type
MB	Symbol
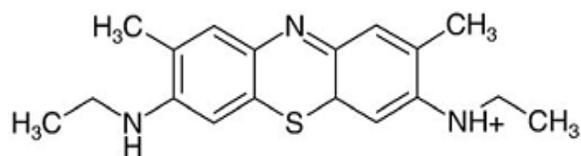
Figure 1) Structural formula of methylene blue dye

Results

Characteristics of adsorbent
SEM are very suitable for study of morphology and characteristics of surface absorbent of fruit Quercus branti tree activvated carbon. Figure 2 showed adsorbent surface properties and morphology before and after MB adsorption on: Quercus branti activated carbon. The results showed that activated carbon from: Quercus branti has a very fine and micro-porosity attributed to the presence of cellulosic materials in absorbent structures, but substances with the lignin structure have coarse porosity (30). Different studies have shown that porosity structure of adsorbent is led to increasing contact surface, amount dye ions adsorption on adsorbent surface, and effective role in adsorption ability (31).
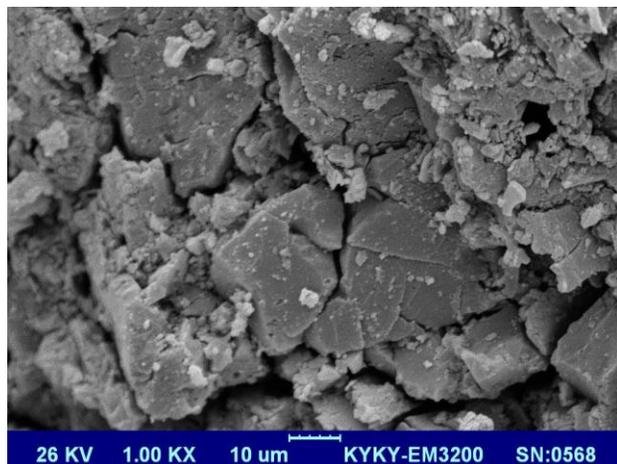
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Figure 2) Picture of (SEM) from Quercus branti activated carbon before adsorption

FT-IR spectra are very important technique for determining the characteristics of functional groups and also their changes in the adsorbents. Change of FT-IR spectra related to adsorbent before and after MB adsorption have shown in Figure 1.a and Figure 1b respectively. In figure 1a has shown a number of adsorption peaks that showed material components. The presence of a Band in 3379.22 board is a reason for stretching frequency of hydroxyl functional group (OH) on the surface of the adsorbent. Adsorption peak in 2920.79 board is related to stretching frequency of the symmetric group-CH_3. Strong bands in 1616.42 board is the sign of stretching frequency of the C=O in carboxylic acid banded with hydrogen of molecular inside is (32). But adsorption peak in 1517.05 board is related to aromatic chain on the adsorbent surface. 1441.28 peak is related to symmetric curve of -CH_3. Frequency, observed peak in around of 1035 may be related to frequency in OH and stretching frequency in C-O-C in adsorbent cellulose structure. In figure 1.b was cleared that after MB adsorption, stretching frequency of group (OH) and also C=O in carboxylic acid with bonded hydrogen of molecular inside was changed and frequencies of 3379.22 and 1616.42 were reduced to 3404.09 and 1602.19 respectively. All changes confirm that MB adsorption on adsorbent was suitable.
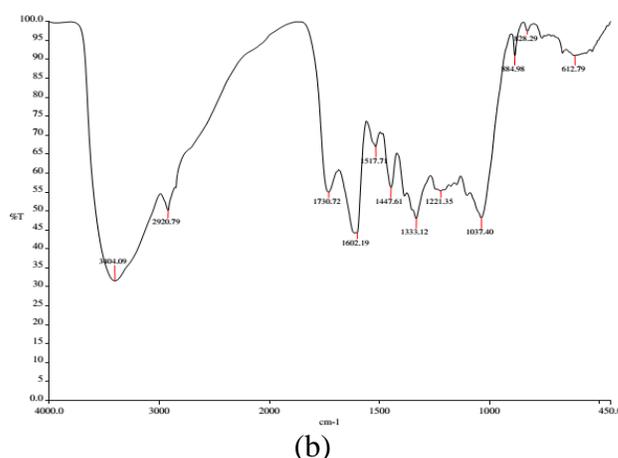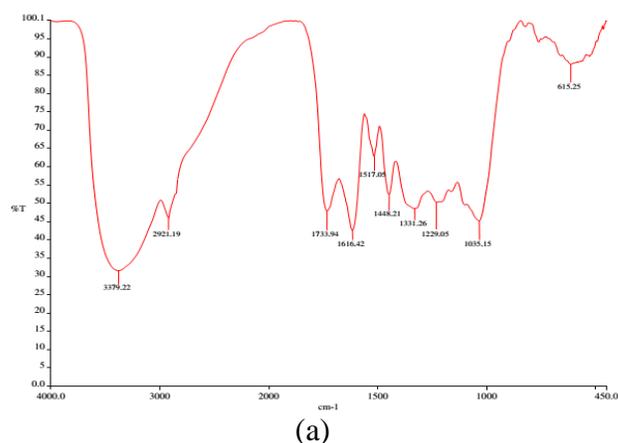

(a)

(b)

Figure 3) FT-IR spectra of Quercus branti activated carbon before (a) and after (b) of MB adsorption respectively.

Obtained results from effect of contact time and initial concentration of methylene blue on efficiency removal have shown in figure 4.
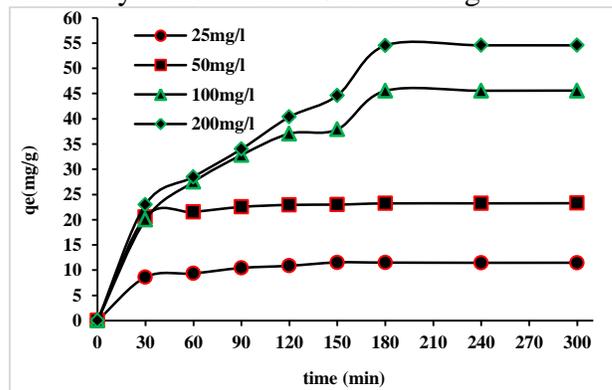

Figure 4) Effect of initial concentration of MB removal and contact time (MB dose= 50,100,200 mg.l^{1-}, adsorbent dose= 2g.l^{-1}, pH=6, reaction (contact) time= 300min)

Obtained results from Effect of pH on MB removal (MB dose =100mg.l^{-1}, adsorbent dose=2mg.l^{-1}, Contact time=180 min) have shown in figure 5.
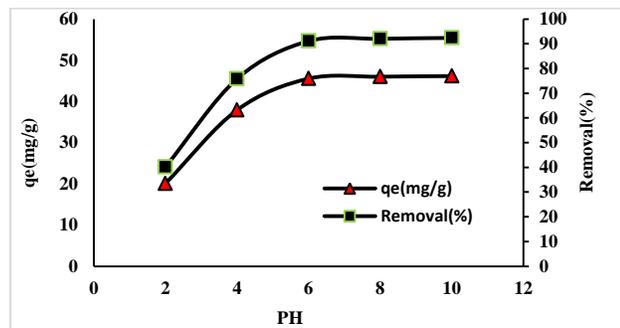

Figure 5) Effect of pH on MB removal (MB dose =100mg.l^{-1}, adsorbent dose=2mg.l^{-1}, Contact time=180 min)

To study the effect of adsorbent dose on the adsorption process methylene blue on Quercus branti activated carbon, 1, 1.5, 2, 2.5 g.l^{-1} adsorbent concentrations were used in optimal condition. According to figure 6 was cleared that with the increase in the amount of absorption of 0.5 to 2.5 g.l^{-1} removal efficiency was increased from 42% to 92%, but absorption capacity was reduced from 84 to 36.5%. Because MB removal in 2g.l^{1-} of adsorbent was 91.08 and was near to 2.5 g.l^{1-}, 2g.l^{-1} was selected as optimal dose.
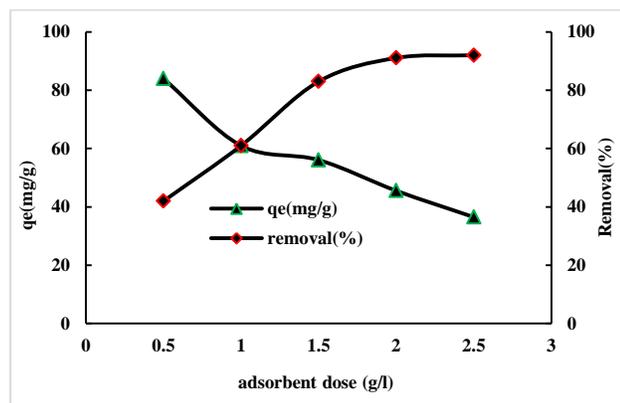

Figure 6) Effect of adsorbent dose on MB removal (MB dose =100 mg.l^{1-}, pH=6, reaction time=180 min

The effect of different temperatures (20, 30, 40, 50 °C) on removal efficiency of MB in optimal condition was shown in figure 7.
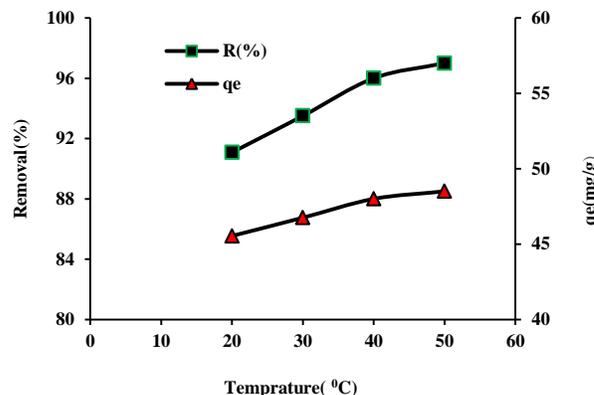

Figure 7) Effect of Temperature on MB removal (MB dose=100mg.l^{-1}, Adsorbent dose=2g.l^{-1}, pH=6,

Discussion

Effect of contact time and initial concentration of methylene blue

Adsorption process of MB consists of two stages. In the first stage, rapid initial adsorption was observed at 30 min and in second stage, adsorption process was slow and was performed during time of 30-180 min. in this stage, the absorption capacity was slower than first stage. It was cleared that increasing time over 180 min had no significant effect on adsorption capacity. It can be stated that in this time with regard to dynamic equilibrium, adsorption and desorption for color are the same. This may be due to the fact that during the adsorption process, the dye molecules by the process of mass transfer are rapidly received to boundary layer .Then they slowly released from the boundary layer into the absorber surface. Because the most active adsorption sites were occupied, the color particles were dispersed into the adsorbent pores (33). Also it was observed that the adsorption capacity increased with increasing the initial concentration of the dye. Mass transfer driving force increased with increasing

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initial dye concentration and also reaction between methylene blue and absorption capacity was increased, therefore adsorption capacity was increased (34). Ahmad et al in 2010 found that Azo dye adsorption amount on granular activated carbon was increased with increasing initial concentration of Azo dye, and this phenomenon was because of increasing concentration gradients (35).

Effect of initial pH

In the process of absorption, pH plays a key role in absorptive capacity and effects on the adsorbent surface features (functional groups on the active sites), degree of ionization and removal efficiency. The effect of these parameters on the adsorption process of MB on activated fruit Quercus branti tree, 100 mg.l^{-1} colors at pH 2-10 and 2g.l^{-1} adsorbent at a speed of 140 rpm for 180 min were studied. Figure 5 Effect of pH on the adsorption capacity and removal efficiency shows. According to this figure, with increasing of pH (from 2 to 10) MB removal will be increased from 40.16 to 92.3%. It was cleared that minimum and maximum of removals belonged to pH 2 and 10 respectively. MB removal percentages in pH=6 and 10 were almost near to gather (91.08 and 92.3% respectively), therefore pH 6 was selected as optimum pH. In this study, decreasing pH has been led to significant reduction of removal efficiency and adsorption capacity. Because there are almost more protons at pH low, reducing the electrostatic force between the dye anions and positive charge of adsorption sites occurs. Hence the ionic repulsion between the positive charge of adsorbent surface and the dye molecules is reduced (36). As is observed from the figure, the color removal rate increases with increasing pH, because of increasing negative charge on adsorbent in this situation (37). The high adsorption capacity observed at high pH values is due to increasing the hydroxyl ions and thus increasing the electrostatic attraction between the positive and negative charges of adsorption sites.

Study of Gercel et al in 2007 showed that maximum MB removal from aqueous solutions on activated carbon prepared from agricultural wastes obtained at pH=6 (38).

In other studies, such as adsorption of MB and malachite green on activated carbon prepared from agricultural wastes (39) and bio adsorption of reactive colors on rice bran from aqueous solutions, efficiency removal was increased by increasing pH. Surrounding of adsorbent surface by carbocyclic groups is led to low efficiency in low pH (19). N. Sivarajasekar et al study about bio adsorption of AZO dye confirms these results (40). In contrary, Hameed et al showed that increasing pH (2 to 13) was caused to decreasing green acid dye 25(AG25) adsorption that its reason was increasing hydroxyle ions and changing adsorbent surface charge (17). According to results of previous studies and this study was cleared that pH effects on pollutions adsorption related to type of pollution and adsorbent were difference and importance.

Effect of the adsorbent dose on methylene blue removal

In description of this fact it can be stated that increasing the amount of adsorbent was led to increasing of active and available sites for reaction processes (41), also unsaturated adsorption sits remained from high amounts of sorbent particles were led to decreasing surface area and increasing path length (42-43). Results of Asgari et al and Amin et al confirm our results (44,29).

Effect of Temperature

According to the results, it was cleared when the temperature is increased from 20 to 50, adsorption capacity and removal efficiency increased. Its reason is that the increase in temperature increases the solubility of the dye and then increases collisions between adsorbent and adsorb and thereby increases the absorption capacity. Also, increasing temperature led to increasing pore size of the adsorbent surface and increasing adsorbent capacity (45-46). Other studies also showed that the adsorption

process can be improved by increasing the mobility of adsorb molecules (47).

Conclusion

Obtained results from MB removal on Quercus branti activated carbon, as adsorbent, in aqueous solutions shown that:

1- MB removal is depended on pH, and at high pH removal amount is more than others.

2- Relation between MB removal with pH, adsorbent dose and reaction time is a direct relation, but with dye concentration is indirection.

3- Optimal condition in this study was obtained in initial dye concentration 100 mg.l^{-1}, pH6, temperature 20 ^{0}C, reaction time 180 min, speed 140 rpm, and adsorbent dose 2g.l^{-1}

4- Maximum MB removal in optimal condition was 91.08%.

5- According to results of this study MB removal by Quercus branti activated carbon as economic, available, costly, and natural adsorbent, is recommended. Also, it seems that oak activated carbon be favorable for other dye pollutions.

Footnotes

Acknowledgments:

Conflict of Interest:

The authors declared no conflict of interest.

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