Original Article

Removal of Reactive Black 5 dye from Aqueous Solutions by Adsorption onto Activated Carbon of Grape Seed

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Abstract

Background and purpose: The control of environmental pollution especially the pollution of water resources is one of the main challenges of researchers throughout the world. So, this study aimed to investigate the efficiency of reactive black 5 dye removal from aqueous solutions by adsorption onto activated carbon of grape seed.

Materials and Methods: At first, the grape seed adsorbents were prepared in three forms of raw, treated by concentrated phosphoric acid, and calcined at 400, 600, and 800 °C. Then, the efficiency of adsorbents to the removal of the Black 5 dye was studied in the concentrations of 100 to 700 mg/Lat pH 2- 10 and 1- 10 g/L of adsorbent during 120 min. The change of concentration was studied via the spectrophotometry procedure at the wavelength of 597 nm. Finally, the Langmuir and Freundlich adsorption isotherm was determined.

Results: The results showed that the dye removal efficiency decreased by increasing pH, and increased by raising the contact time and the amount of adsorbent. So, in this process, the highest percentage of Black 5 dye removal (83.08%) was obtained at pH 2 and t 120 min using the raw adsorbent, 200 mg/L initial concentration of dye, and 10 g/L of carbon. The process considerably followed the Langmuir adsorption isotherm (R^2 0.999).

Conclusion: The grape seed was found to have the highest level of efficiency in dye removal, and according to the availability of adsorbent and its low price, this method could be used as an applicable procedure for the removal of Black 5 dye from aqueous solutions.

Keywords: React Black 5; Dye Removal; Grape seed; Adsorption; Activated carbon

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1. Introduction

The increase of dye production and its application around the world results in producing wastewater with the high pollution which causes concerns in the case of creating environmental pollutions (1). The dye wastewater is produced in different industries, such as textile and dyeing industries, pharmaceutical industry, food industry, production of cosmetics, and paper and leather industry (2). The dye wastewater discharge leads to the undesirable appearance, reducing water usage for the urban, agricultural, and industrial uses, and the inappropriate environment for the recreational uses. Also, the presence of aromatic rings in these dyes has the carcinogenicity and mutagenicity effects and threats the human and animal's life (3). About 12% of dye used in the textile industry enters wastewater, of which 20% is discharged into environment without purification (4). In recent years, industries have been under the pressure of authorities and public opinions to purify the wastewater before discharging into the environment. The reactive Black dye is one of the most common dyes used in the textile industry which consists of a diazo moiety and four phenolic groups, and is decomposed into sulfonate anions after being dissolved in aqueous solutions. The reactive Black dye produces the dark blue dye after dissolving which is due to the aromatic rings linked to the azo groups (5). Therefore, the removal of dye from these wastewaters is essential. By the development of textile dye productions, the new dyes are introduced which have the better dyeing properties but this quality improvement results in the stability of dye structures and increases their resistance against the biodegradation and chemical processes. The majority of dyes used in textile industries are non-biodegradable due to the formation of strong complexes, and the common processes of wastewater purification, such as coagulation, flocculation, and chemical precipitation, are not considered as effective removal methods (6). In addition, these techniques have further limitations and problems, such as high costs, production of sludge, and insufficient efficiency (7). Among these processes, the adsorption method is an environmentally friendly, simple, and economic procedure which has widely been considered in the recent decades. However, the main challenge related to the adsorption process is finding a low-cost adsorbent. To reduce the cost of preparation, using low-cost materials, such as agricultural crop and industrial residues, is mainly considered as a potential option for producing activated carbon (8). The grape seed is known as a low-cost, natural, and available adsorbent, and has been used in some research for the removal of pollutants. In 2012, M. Al Bahri et al. used the activated carbon, prepared from the grape seed after the chemical activation by phosphoric acid, for the removal of diuron from water (9). In 2009, Didem Ozcimen applied the activated carbon of chestnut bark and the grape seed for the removal of copper from aqueous solutions (10). The purpose of this research was investigating the efficiency of activated carbon obtained from the grape seed in the adsorption of Black 5 dye from the synthetic water samples.

2. Material and methods

All chemicals, such as the reactive Black 5 dye $C_{26}H_{21}N_5Na_4O_{19}S_6$ (RB_5), sodium hydroxide, and hydrochloric acid were purchased from the Merck Company, Germany. The double distilled water was used in all steps. The adjustment of pH was done *via* the Denver Ultra basic-UB10 made in America device using hydrochloric acid and sodium hydroxide 1 N (11). To prepare the adsorbent, the grape seed was used as raw, treated by the concentrated phosphoric acid in the proportion of 3:1, and also calcined at 400, 600 and 800 °C. To remove the pollutions, and especially the dyes, first, the grape seed was boiled in boiling water till the dye disappeared, and dried at 120 °C for 24 hours to be used in the carbonization step. After determining the initial structural properties of the grape seed (carbon, nitrogen, hydrogen, sulfur, and volatiles), it was heated by an electric furnaces (a horizontal type of Alfa model) at 0, 300, 500, and 800 °C (the temperature should already be reached to the desired level) for 60 min. The produced carbon was weighed for determining the humidity percentage and the amount of obtained adsorbent, and kept into the sealed container of desiccator in a cool place.

The dye used in this research is commercially named as the reactive Black 5 (RB_5) with the molecular weight of 991.82 from the diazo dye groups, which is widely used in paper and textile industries. Its maximum light absorption spectrum is at 597 nm (5). The solution of reactive Black 5 with the concentration of 1000 mg/L was prepared using the distilled water, and the stock solution was kept in the refrigerator to prevent the concentration changes. Using the stock solution of Black dye, the determined concentrations (100, 200, 300 and 700 mg/L) were prepared, and then the effects of pH and the amount of adsorbent were studied regarding the achievement of the highest efficiency. The solution pH was adjusted in the range of 2 to 10 using nitric acid and sodium hydroxide 0.1 M. After setting the experimental conditions, adsorbent (10 g) was added to the 1000 mL of solution containing the determined concentrations of dye, and then reacted on the shaker with the rate of 100 rpm for 120 min. After 12 min, the samples were filtered using the Whatman filter paper with the thickness of 42 μm, and the concentration of dye remained in the solution was characterized by the UV-Vis spectrophotometer method at the wavelength of 597. Also, the removal percentage was calculated through the following formula.

$$\%\,Removal = \frac{C_0 - C_f}{C_0} \times 100$$

At the end, about 100 samples were evaluated through double test based on one factorial method, where C_0 is the initial concentration of dye, and C_f is the concentration of dye remained after the adsorption.

The results were then keyed in a table using Microsoft Excel Software-2013 so as to determine the highest removal percentage, and the optimum parameters were then reported. To specify the maximum adsorption and determine the optimum conditions for the optimal adsorption, the Langmuir and Freundlich adsorption isotherm was performed. The Langmuir model is one of the most significant one-layer adsorption models, and is based on a fixed number of adsorption sites in which each site has the adsorption ability of one molecule. All sites are in equal conditions, and there is not any connection between the adsorbed molecules. The Langmuir model equation is as follows:

$$\frac{1}{q} = \frac{1}{q_m.b}\left(\frac{1}{C}\right) + \frac{1}{q_m}\text{Ce}$$

Here, q_m is the maximum adsorption capacity (mmol/g), and b is the Langmuir adsorption constant. To calculate these two parameters, the C_e/q_e versus Ce graph should be drawn.

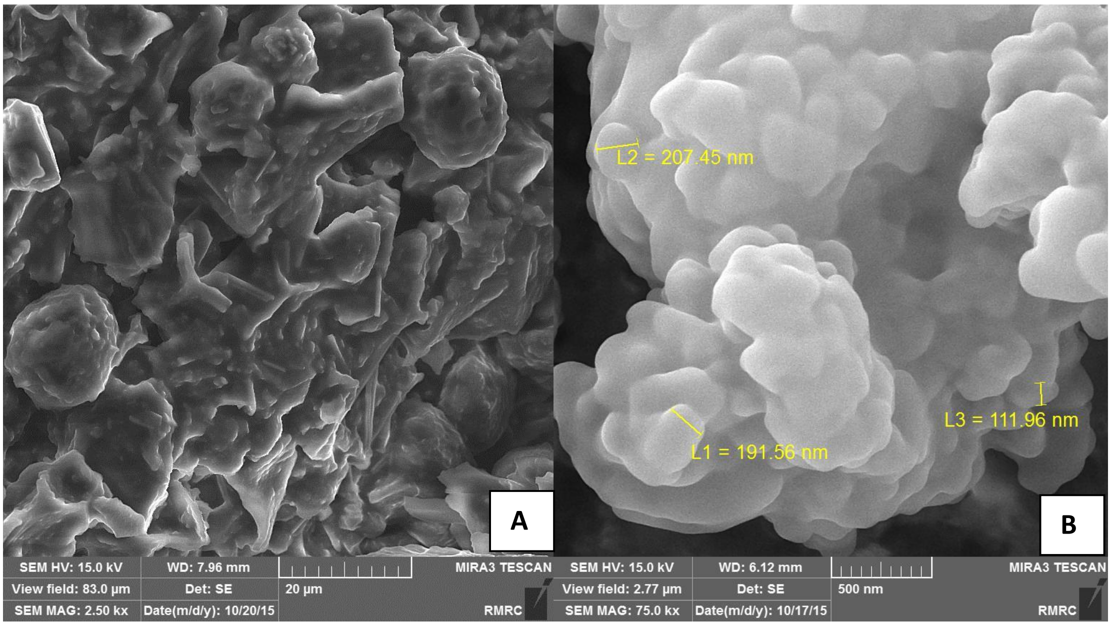
Another isotherm adsorption model is the Freundlich model. The Freundlich model is based on one-layer adsorption on the heterogeneous adsorption sites with unequal and opposite energies. In Freundlich model, when K_f increases, the adsorption capacity of adsorbent rises. Also, the amount of n between 1 and 10 illustrates the appropriate adsorption process as follows:

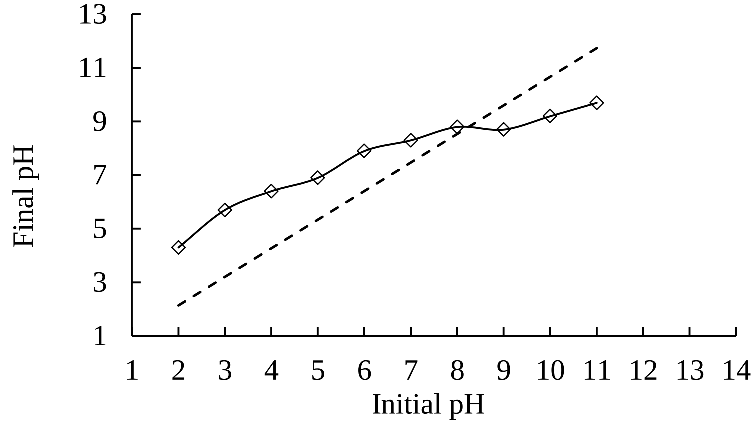
$$\log q_e = \log K_F + \frac{1}{n}\log Ce$$

In this formula, n and K are the Freundlich constants. This model can be easily introduced by drawing the logarithmic and linear form (11).

3. Results

Due to the high efficiency of raw grape seed adsorbent – rather than the other types of adsorbents – that was produced and used in this sudy, the surface morphology of raw grape seed was characterized using SEM micrograph, as shown in Figures 1A and B. The SEM micrograph, shows that it is Smooth and uniform surface and Non-porous. The BET surface area of grape seed was measured by means of standard BET equation, of which its value is 623 m^2/g and mesopore volume (0.17 cm^3/g). The pHpzc "pH drift" of grape seed adsorbent was also determined to be 8.1, signifying a positive surface charge for a solution with a pH below 8.1 and a negative surface charge for a solution with a pH greater than 8.1, as shown in Figure 1C (12-15). Figure 1D also illustrates the FTIR spectrum of grape seed adsorbent, including the peak wave numbers and the corresponding assigned groups. This result implies that a complex grape seed adsorbent surface is involved in adsorbing dye (16-18).





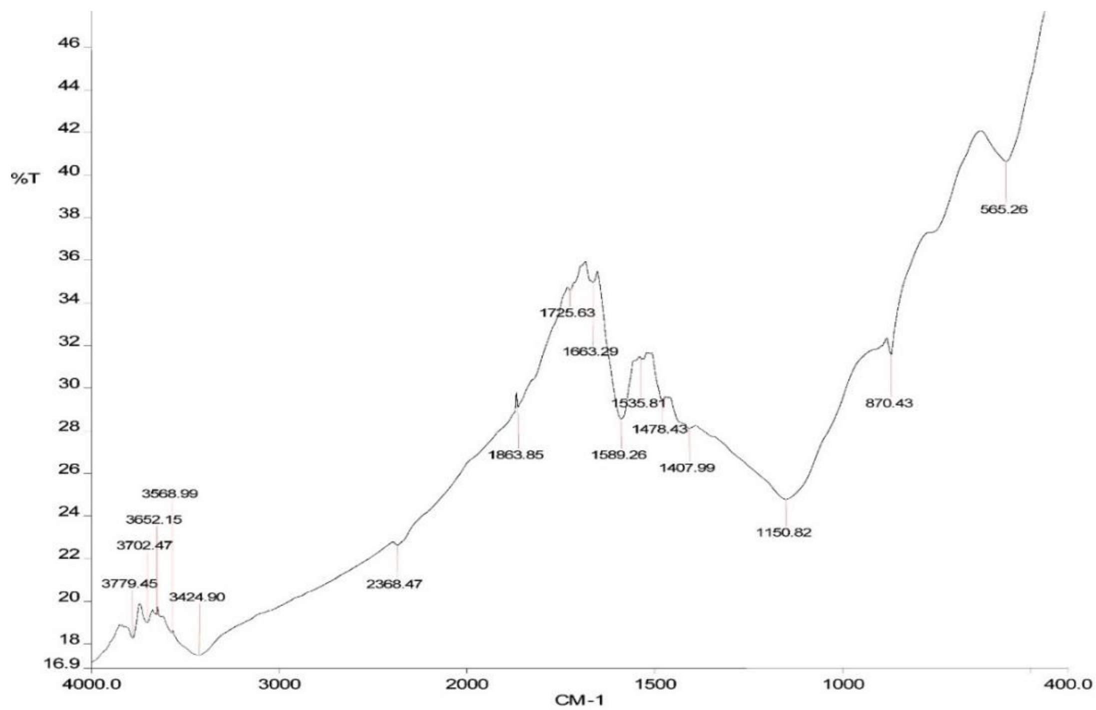


Figure 1. A: SEM micrographs of Raw of Grape Seed befor dye adsorption **B:** SEM micrographs of Raw of grape seed after dye adsorption **C:** Determination of pHpzc **D:** FTIR spectrum of Activated Carbon of grape seed at wave numbers from 400 to 4000 cm^{-1}

Figure 2 depicts the effects of varying adsorbent types (raw, acidic treated, and calcined adsorbents) on the dye adsorption by the grape seed at different temperatures. Figure 2 shows that the acidic treated and calcined adsorbents did not have great impact on the dye adsorption by the grape seed, and the highest percentage of adsorption (51.97%) was achieved when the raw adsorbent was used.

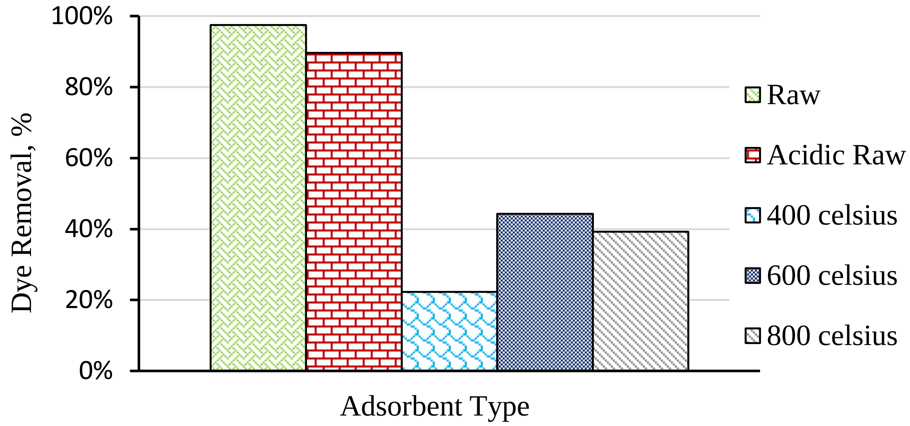


Figure 2. The removal yield of Black 5 dye by the raw and modified grape seed (200 mg/L dye concentration, pH 2, and t 120 min)

Figure 3 shows the effects of pH changes on the adsorption of Black dye in the pH range of 2-10. As illustrated in the figure, a considerable increase in the pH level from 2 to 3 decreased the dye adsorption, leading the decreasing trend to be mild. Whereas, after reaching neutral pH, a slight increase in pH level reduced the adsorption. As a result, the highest removal percentage (83.08%) was obtained at pH 2.

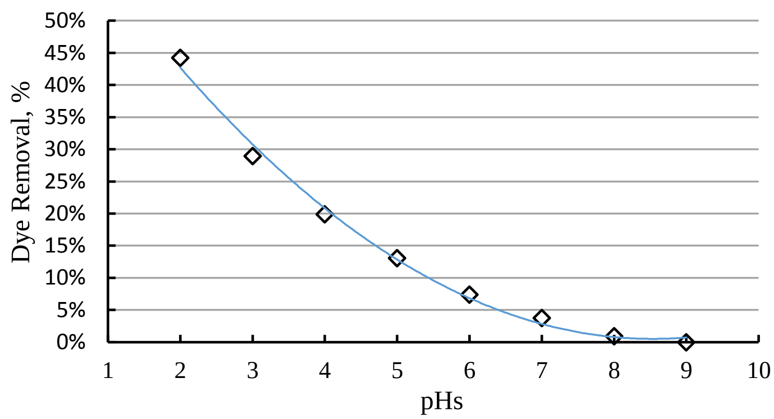


Figure 3. The effect of pH on the adsorption of Black dye by the grape seed (200 mg/L dye concentration and t 120 min)

Figure 4 depicts the effect of adsorbent amounts on the adsorption of Black 5 dye. As shown in the figure, increasing the amount of adsorbent from 1 to 10 g/L raised the dye removal efficiency, and the concentration of remained dye decreased from 71.69 to 33.84. Therefore, the highest removal yield (83.08%) was achieved using the 10 g/L optimal amount.

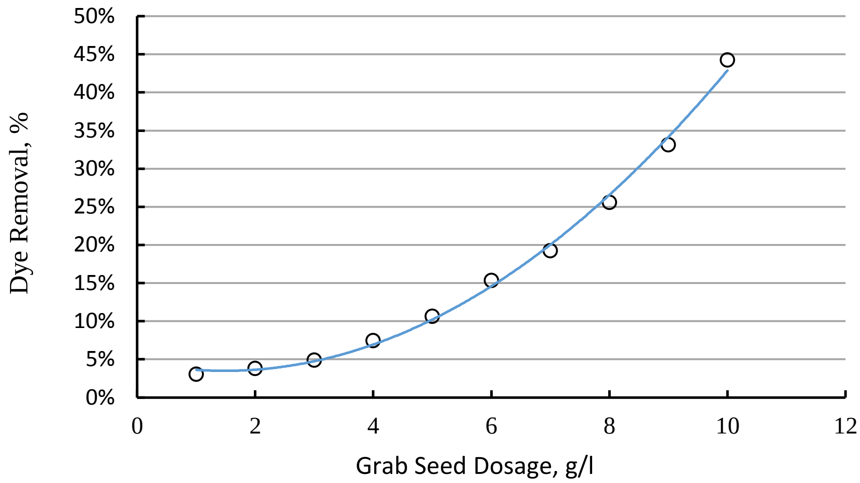


Figure 4. The effect of adsorbent amount on the adsoption of Black dye by the grape seed (200 mg/L dyedye concentration, t 120 min, pH 2)

Figure 5 shows the effect of initial concentration of dye on the adsorption rate. As depicted in the figure, raising the initial concentration of dye from 100 to 700 mg/L reduced the dye removal yield, so that the adsorption percentage of 100-300 mg/L was between 77-87%, while in higher concentrations, the adsorption rate decreased and reached 48.97% in the concentration of 700 mg/L.

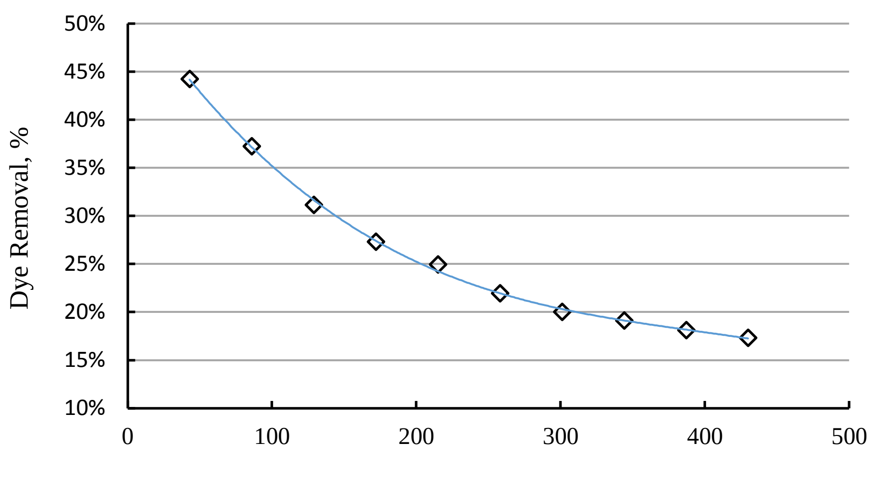


Figure 5. The effect of initial concentration of dyedye on the adsorption of Black 5 dye (pH 2 and t 120 min)

Figure 6, on the other hand, shows the effect of contact time on the dye adsorption rate. As illustrated in it, increasing the contact time raises the dye removal yield. At t = 120 min, adsorption yield of the Black dye was found to be about 83.08%, resulting in the optimal contact time to become 120 min.

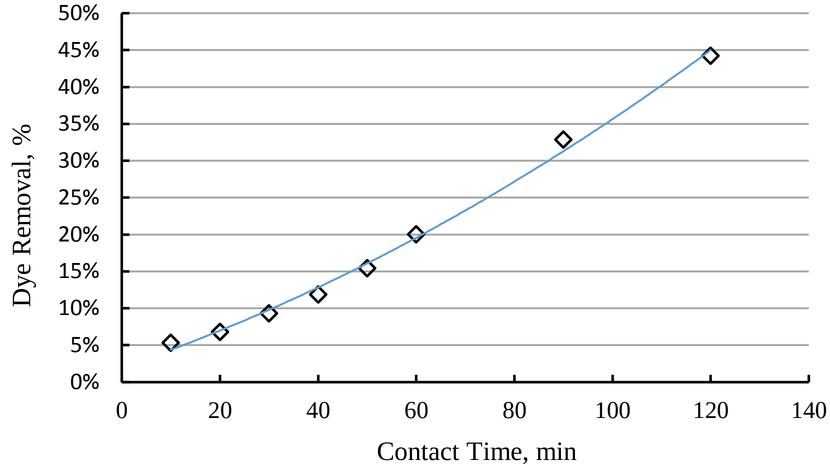


Fig 6: The effect of contact time on the adsorption of Black 5 dye (200 mg/L dye concentration and pH 2)

Figure 7 depicts the effect of varying liquid temperature on the dye adsorption rate. As shown in the figure, in the current study, decreasing temperature raised the dye removal yield, so that reducing temperature from 50 to 27 °C increased the dye adsorption yield from 33.40% to 83.08%.

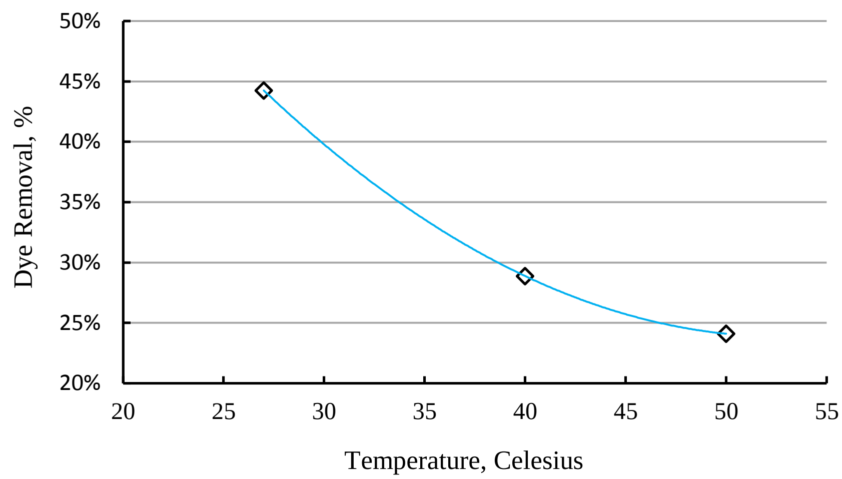


Figure 7. The effect of temperature on the adsorption of Black 5 dye (200 mg/L dye concentration, t 120 min, pH 2)

Figures 8 and 9 illustrate the amount of substance adsorbed at constant temperature. According to the regression coefficients (R^2) of these two adsorption models which are 0.999 and 0.9291 for the Langmuir and Freundlich isotherms, respectively, it can be concluded that the adsorption of Black 5 dye by the grape seed follows the Langmuir adsorption isotherm considering the higher level of correlation coefficient.

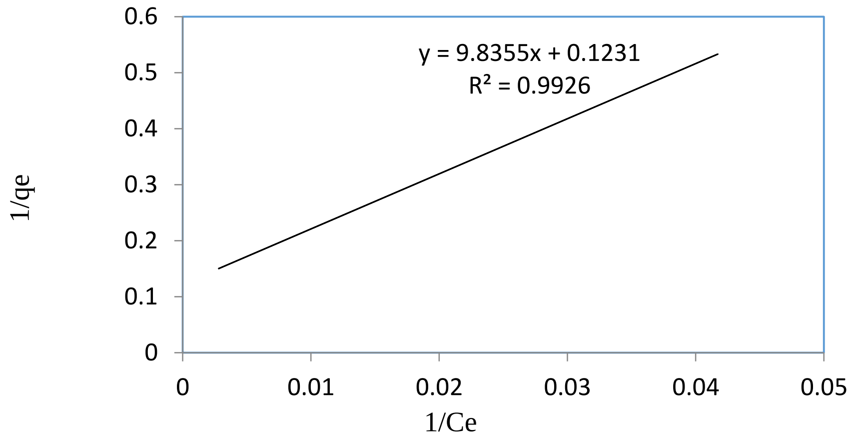


Figure 8. The Langmuir isotherm for the adsorption of Black 5 dye by the grape seed (pH = 2 and t = 120 min)

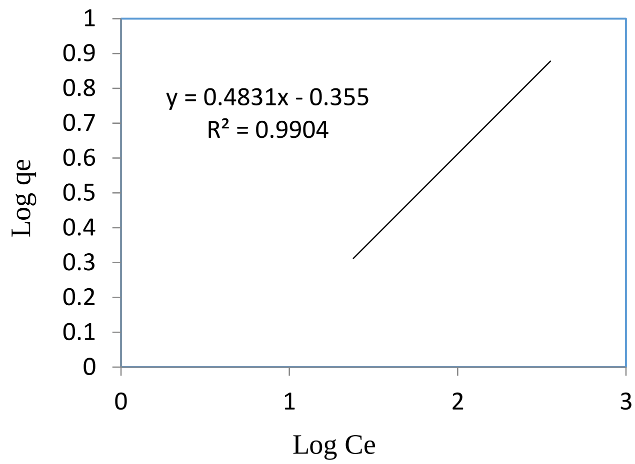


Figure 9. The Freundlich isotherm for the adsorption of Black 5 dye by the grape seed (pH 2 and t 120 min)

4. Discussion

The results of the present study revealed that the pH of dye wastewater is greatly effective on the dye adsorption and the adsorption capacity. This issue is caused by the changes of functional group ionization in the adsorption sites and finally the charge changes of adsorbent surface and ionization of substances present in the solution (19). Figure 3 shows the effect of solution pH on the adsorption capacity of activated carbon prepared from the grape seed. As depicted in the figure, the Black 5 dye was removed from the aqueous solutions at acidic pH with the high yields, so that the adsorption decreased from 83.08 to 58.97% by increasing pH from 2 to 10. In 2015, Narooie *et al.* evaluated the efficiency of ashes from palm and pistachio wastes in removal of reactive red 120 dye from aqueous, and reported that increasing pH decreased the removal efficiency. They found that this issue can be due to the close connection between the bonds of adsorbent site and hydrogen ions at acidic conditions which acts as a ligand bond between the adsorbent surface and dye molecules (18). In Jaqueline Moura *et al.* compared the different physical forms of chitosan for the removal of reactive Black 5 from aqueous solutions, and reported that increasing pH decreased the removal yield, hence by reducing pH from 8 to 4, the yield reached about 99%. They believed this event was due to an increase in the level of H^+ ions in the solution which could then facilitate the conversion of R-NH_2^+ to R-NH_3^+ (20). Dotto et al. evaluated the adsorption rate of Reactive Black 5 on chitosan-based materials, and reported that lower pH reactive black 5 adsorption was done better than higher pH (21). Zandi Pak and Ardakani investigated the removal of anion anionic dyes (blue 106 and green acid 25) by the oxidized multi-walled carbon nanotubes, and reported that increasing pH from 2 to 7 decreased the removal yields, so that at pH 2, the yields of 95% and 89% were achieved for the blue 106 dyes and green acid 25 adsorption, respectively (22). Also, the solfunate functional group presented in the dye was deionized in the water to create the anionic form of the molecule. On the other hand, raising pH resulted in increasing negative charge of adsorbent, and repulsive forces prevented the adsorption of dye by the adsorbent. In this study, the highest yield was achieved in acidic pH, which could then be in agreement with the results of the aforementioned studies. So, the pH 2 was considered as the optimum pH.

The amount of adsorbent is one of the most important parameters in the adsorption process, and plays a key role in the adsorption of pollutants (5). Figure 4 shows the effect of adsorbent concentration on the removal of dyes in the constant concentration of 200 mg/L. As depicted in the figure, increasing the amount of adsorbent raises the dye removal yield. In this case, raising concentration from 1 to 10 g/L increases the removal yield from 64.15 to 83.08%. In 2012, Gholami et al. studied the removal of Black 22 from the aqueous solutions by the activated carbon prepared from the orange peel, and reported that increasing the amount of adsorbent from 0.1 to 1 g/L raises the removal yield from 87.1 to 95.2% (23). Rahmani et al. in 2012 removed the reactive Black 5 from the aqueous solutions using the adsorption on the strong basic anionic ion exchange resins. As they reported, raising the initial concentration of adsorbent from 0.2 to 1 g/100c increased the removal efficiency from 65.34 to 99.99% (6). In 2006, Eren and Nuran Acar used the powder of activated carbon and Afsin-Elbistan ash to remove the Black 5 dye from the aqueous solutions. They reported that the increase of adsorbent amount raised the removal yield which was due to the increase of active or available surface of the adsorbent (16, 25). In the current study, the highest yield was achieved in higher concentrations, which was then in agreement with the above research. Hence, the initial concentration of 10 g/L was chosen as the optimum concentration.

In the adsorption process, the initial concentration of ions in the solution plays a key role as a driving force to overcome the resistance of mass transfer between the liquid and solid phase (5). Figure 5 depicts the initial concentration changes of dye solutions for the adsorption by the grape seed in the range of 100-700 mg/L. As illustrated in the figure, reducing the dye concentration increased the adsorption yield. In 2013, Yousefi *et al.* investigated the efficiency of modified wheat straw in the removal of reactive Black 5 from the aqueous solutions, and reported that increasing the dye initial concentration from 10 to 100 mg/L decreased the dye removal yield from 100 to 77.99%, while the amount of dye adsorbed raised from 2 to 15.58 mg/g. According to their research findings, this reduction in removal yield could be due to a decrease in adsorption sites in comparison with the entrance dye molecules (5). In 2012, Rahmani *et al.* used the strong basic anionic ion exchange resins to remove the reactive Black 5 from the aqueous solutions. As they reported, increasing the dye initial concentration decreased the removal efficiency, hence the increase of concentration from 50 to 300 mg/L resulted in a reduction in the removal efficiency from 99.97 to 24.40% (6). In addition, it was revealed that in higher concentrations, the molecules competed with each other to reach the adsorption sites, and the adsorption rate decreased. In this study, the highest yield was achieved in lower concentration which could then again be in agreement with the results of above-mentioned research. Therefore, the initial concentration of 200 mg/L with the yield of 83.08% was considered as the optimal concentration.

The contact time is one of the effective factors on the adsorption process (15). Figure 6 shows the effect of optimum contact time on the Black 5 adsorption at pH 2 in the concentration of 200 mg/L. As depicted in Figure 6, the increase of contact time raises the dye removal yield. Özçimen and Ersoy-Meriçboyu in 2009 studied the removal of copper from the aqueous solutions using the activated carbon obtained from chestnut bark and the grape seed. They reported that raising the contact time increased the removal yield. They also found that the time needed for reaching the equilibrium was 90 and 120 min for the chestnut bark and the grape seed, respectively (10, 13). In 2013, Ghaneian *et al.* investigated the efficiency of flower powder of crap plant in the reactive Blue 19 removal from the synthetic wastewater. They reported that increasing contact time raised the dye removal. In this case, the highest dye removal yield was obtained in the first 30 min, which was then due to a decrease in solution concentration and the active sites on the adsorbent surface (16, 26). In 2016, Mousavi *et al.* studied the isotherm adsorption and the effective factors on the methylene blue removal using the activated carbon prepared from the grape leaves. They reported that increasing the contact time from 10 to 110 min raised the dye removal percentage (14, 26). Furthermore, an increase in contact time raised the collision chance of pollutants with the adsorbent, leading to an increase in the pollutant adsorption by the adsorbent. In this research, the yield of 83.08% was achieved at t = 120 min which is also consistent with the results of the aforementioned studies. So, the contact time of 2 h was chosen as the optimum contact time.

As depicted in the Figure 7, decreasing the temperature raises the adsorption. In 2015, Gholinejad *et al.* performed the Ag/CMK-3 nanocomposite to remove the methyl orange G. Based on their results, increasing the temperature from 30 to 60 °C reduced the adsorption on the surface of nanoadsorbent. They reported that this reverse effect was due to the exothermic nature of adsorption reaction (17, 27). In addition, it was revealed that an increase in temperature not only weakened the physical bonds between the dye and carbon adsorbents, but also raised the dye solubility. As a result, the physical bonds between the dye molecules and water were stronger than their bonds with the adsorbent surface. So, the adsorption decreased by increasing temperature. In this study, the highest removal yield was obtained at 27 °C, which is a result in line with the results of the abovementioned research.

The adsorption isotherms depict the molecular part of the adsorbent in the equilibrium condition between the liquid and solid phase. These isotherms describe the behavior of adsorbed part and the adsorbent, and also provide the most important adsorption model (1). In Figures 8 and 9, the Langmuir and Freundlich adsorption isotherm models have been investigated. It is illustrated that the dye adsorption follows the Langmuir isotherm.

Khalid Z. Elwakeel removed the reactive Black 5 from the aqueous solutions using the magnetic chitosan resin, and reported that the dye adsorption followed the Langmuir isotherm (28). In 2013, Radaie *et al.* studied the reactive Blue 19 dye adsorption using the activated carbon prepared from the pomegranate waste, and achieved the highest amount of correlation coefficient (R^2 0.955) (29). In another study conducted by Ghaneian *et al.* in 2014, the powder of jujube stems were used to remove the reactive Blue 19 dye, resulting in the fact that dye adsorption follows the Langmuir isotherm, hence the R^2 was obtained about 0.9 and 0.91 in the concentrations of 25 and 50 mg/L, respectively (30). In the present research, the adsorption of reactive Black 5 dye using the grape seed followed the Langmuir isotherm with the regression coefficient of 0.999 which was then in agreement with the results of above studies.

The main aim of the current research was to investigate the removal efficiency of Black 5 dye using the grape seed *via* the adsorption process. The results showed that the optimum conditions were obtained when 10 g/L adsorbent and 200 mg/L dye were used at 27 °C and pH = 2 during 120 min to furnish the yield of 83.08%. Also, the results depicted that the adsorption of Black 5 dye using the desired adsorbents has been influenced by different parameters, such as pH, the amount of adsorbent, the initial concentration of dye, the contact time, and the temperature, hence increasing the amount of adsorbent and contact time was documented to raise the removal yield while raising pH, the initial concentration of dye and the temperature reduced the removal efficiency. The findings also revealed that the results of adsorption equilibrium follow the Langmuir adsorption isotherm (R^2 0.999). According to the granulated structure of grape seed, the raw form was then found to be an appropriate and low-cost adsorbent in the adsorption of Black 5 dye from the aqueous environments.

Acknowledgments

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Conflict of interest

The authors declare that there is no conflict of interests.

Ethical Issues

Ethical issues have entirely been considered by the authors. The authors have also desperately tried to avoid plagiarism.

Author's contributions

Afsharnia was statistical consultant and text editor of the study. Biglari was designer, performer and leader of the study. Zabihi was advisor of experiment, sampling and analyzing. Javid conducted the study.

References

1. Altıntıg E, Altundag H, Tuzen M, Sarı A. Effective removal of methylene blue from aqueous solutions using magnetic loaded activated carbon as novel adsorbent. Chemical Engineering Research and Design. 2017; 122: 151–163. DOI: 10.1016/j.cherd.2017.03.035
2. Yari AR, Majidi G, Reshvanloo MT, Nazari S, Emami Kale Sar M, Khazaei M, et al. Using Eggshell in Acid Orange 2 Dye Removal from Aqueous Solution. Iranian Journal of Health Sciences. 2015;3(2):38-45. DOI: 10.7508/ ijhs.2015.02.006
3. Khosravi R., Hossini H., Heidari M., Fazlzadeh M., Biglari H., Taghizadeh A and et al. Electrochemical decolorization of reactive dye from synthetic wastewater by mono-polar aluminum electrodes system. International Journal of Electrochemical Science. 2017; 12(6): 4745-4755. doi: 10.20964/2017.06.75
4. Hamzeh Y, Izadyar S, Azadeh E, Abyaz A, Asadollahi Y. Application of Canola Stalks Waste as Adsorbent of Acid Orange 7fromAqueous Solution. Iranian Journal of Health and Environment. 2011;4(1):49-56.
5. Yousefi N, Fatehizadeh A, Ahmadi A, Rajabizadeh A, Toolabi A, Ahmadian M. The efficiency of modified wheat brad in reactive black 5 dye removal from aqueous solutions. Journal of Health and Development. 2013;2(2):157-69.
6. Rahmani AR, Asgari G, Farrokhi M. Removal of Reactive Black 5 (RB5) Dye from Aqueous Solution using Adsorption onto Strongly Basic Anion Exchange Resin: Equilibrium and Kinetic Study. Iranian Journal of Health and Environment. 2013;5(4):509-18.
7. Kannan N, Karthikeyan G, Tamilselvan N. Comparison of treatment potential of electrocoagulation of distillery effluent with and without activated Areca catechu nut carbon. Journal of Hazardous Materials. 2006;137(3):1803-9. PMID: 16842910 DOI: 10.1016/j.jhazmat.2006.05.048
8. Liu H-L, Chiou Y-R. Optimal decolorization efficiency of reactive red 239 by UV/ZnO photocatalytic process. Journal of the Chinese Institute of Chemical Engineers. 2006;37(3):289-98.
9. Al Bahri M, Calvo L, Gilarranz M, Rodriguez J. Activated carbon from grape seeds upon chemical activation with phosphoric acid: Application to the adsorption of diuron from water. Chemical Engineering Journal. 2012; 203:348-56.
10. Özçimen D, Ersoy-Meriçboyu A. Removal of copper from aqueous solutions by adsorption onto chestnut shell and grapeseed activated carbons. Journal of Hazardous Materials. 2009;168(2):1118-1125. PMID:19342167 DOI:10.1016/j.jhazmat.2009.02.148
11. Chen S, Yue Q, Gao B, Li Q, Xu X. Removal of Cr (VI) from aqueous solution using modified corn stalks: Characteristic, equilibrium, kinetic and thermodynamic study. Chemical Engineering Journal. 2011;168(2):909-917. http://dx.doi.org/10.1016/j.cej.2011.01.063
12. Karimaei M., Sharafi K., Moradi M., Ghaffari H.R., Biglari H., Arfaeinia H and et al. Optimization of a methodology for simultaneous determination of twelve chlorophenols in environmental water samples using: In situ derivatization and continuous sample drop flow microextraction combined with gas chromatography-electron-capture detection. Analytical Methods. 2017; 9(19):2865-2872. DOI:10.1039/C7AY00530J
13. Saeidi M., Biglari H., Rahdar S., Baneshi M.M., Ahamadabadi M., Narooie M.R., et al. The Adsorptive Acid Orange 7 using Kenya tea Pulps Ash from Aqueous Environments. Journal of Global Pharma Technology. 2017;9(4):13-29.

14. Rahdar S., Ahamadabadi M., Khaksefidi R., Saeidi M., Narooie M.R., Salimi A., et al. Evaluation of Phenol Removal from Aqueous Solution by Banana Leaf Ash. Journal of Global Pharma Technology. 2017;9(3):20-8.
15. Baneshi MM, Naraghi B, Rahdar S, Biglari H, Saeidi M, Ahamadabadi M., et al. Removal of remazol black B dye from aqueous solution by electrocoagulation equipped with iron and aluminium electrodes. Iioab journal. 2016;7:529-35.
16. Rahdar S, Khaksefidi R, Alipour V, Saeidi M, Narooie MR, Salimi A., et al. Phenol adsorptive by cumin straw ash from aqueous environments. Iioab journal. 2016;7:536-41.
17. Khaksefidi R, Biglari H, Rahdar S, Baneshi M.M., Ahamadabadi M., et al. The removal of phenol from aqueous solutions using modified saxaul ASH Research Journal of Applied Sciences, 2016; 11 (11):1404-1410. DOI: 10.3923/rjasci.2016.1404.1410
18. Narooie M.R., Rahdar S, Biglari H, Baneshi M.M, Ahamadabadi M, Saeidi M., et al. Evaluate the efficiency of ashes from palm and pistachio wastes in removal of reactive red 120 dye from aqueous. Research Journal of Applied Sciences, 2016; 11 (11):1411-1415. DOI: 10.3923/rjasci.2016.1411.1415
19. Zazouli M, Balarak D, Mahdavi Y, Ebrahimi M. Adsorption Rate of 198 Reactive Red Dye from Aqueous Solutions by using Activated Red Mud. Iran J Health Sci. 2013; 1 (1): 36-43. DOI: 10.18869/acadpub.jhs.1.1.36
20. Moura JM, Gründmann DD, Cadaval TR, Dotto GL, Pinto LA. Comparison of chitosan with different physical forms to remove Reactive Black 5 from aqueous solutions. Journal of Environmental Chemical Engineering. 2016;4(2):2259-2267.21. Doi: 10.1016/j.jece.2016.04.003
21. Dotto GL, Ocampo-Pérez R, Moura JM, Cadaval TR, Pinto LA. Adsorption rate of Reactive Black 5 on chitosan based materials: geometry and swelling effects. Adsorption. 2016; 22(7):973-983. Doi:10.1007/s10450-016-9804-y
22. Sobhanardakani S, Zandipak R. Removal of Anionic Dyes (Direct Blue 106 and Acid Green 25) from Aqueous Solutions Using Oxidized Multi-Walled Carbon Nanotubes. Iranian Journal of Health Sciences. 2015;3(3):48-57. DOI: 10.7508/ijhs. 2015. 03.006
23. Gholami H, Gholami M, Gholizadeh A, Rastegar A. Use of orange peel ash for removal of direct black 22 dye from aqueous environments. Journal of north khorasan university of medical sciences, 2012; 4(11):45-56.
24. Eren Z, Acar FN. Adsorption of Reactive Black 5 from an aqueous solution: equilibrium and kinetic studies. Desalination. 2006; 194(1):1-10. https://doi.org/10.1016/j.desal.2005.10.022
25. Ghaneian M, Dehvari M, Jourabi Yazdi N, Mootab M, Jamshidi B. Evaluation of efficiency of Russian Knapweed flower powder in removal of Reactive Blue 19 from synthetic textile wastewater. Journal of Rafsanjan University of Medical Sciences. 2013;12(10):831-842.
26. Mousavi SA, Khashij M, Shahbazi P. Adsorption Isotherm Study and Factor Affected on Methylene Blue Decolorization using Activated Carbon Powder Prepared Grapevine Leaf. Safety Promotion and Injury Prevention. 2016;3(4):249-56.
27. Jafar Gna, Torkian L, Daghighi Am. Investigation of orange g adsorption by ag/cmk-3 nano composite. 2015; 9(2): 47-54
28. Elwakeel KZ. Removal of Reactive Black 5 from aqueous solutions using magnetic chitosan resins. Journal of hazardous materials. 2009; 167(1):383-392. PMID: 19272711DOI:10.1016/j.jhazmat.2009.01.051
29. Radaei E, Moghadam SA, Arami M. The Study of the Adsorption of Reactive Blue 19 Dye by Activated Carbon from Pomegranate Residue. Journal of Water & Wastewater. 2014; 25(92):27-34.
30. Ghaneian MT, Ehrampoush MH, Sahlabadi F, Mootab M, Rezapour I, Jasemizad T. Reactive blue 19 dye adsorption behavior on Jujube stems powder from synthetic textile wastewater: Isotherm and Kinetic adsorption studies. J Community Health Res. 2014; 3(1):67-78.