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Determination of Optimum Exposure Time and pH for dye Adsorption using an Adsorbent made of Sewage Sludge

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ABSTRACT: In this research feasibility study of using sewage sludge as an adsorbent for the removal of Acid Blue 25 dye was carried out. Sewage sludge was first dried in an oven at 105° C for 24 hours. Then the dried sludge was washed with HCl (98%) for two hours and then with distilled water to a pH of approximately neutral. Then, it was carbonated and activated in a Nitrogen-based electric furnace at 600C° for two hours. Molecular Structure of the adsorbent was determined through XRD technique. The results of XRD revealed that the adsorbent produced for this study was mainly composed of SiO_2. The experiments were carried out at pH of 3, 5, 7, 9, and 11 and for exposure times of 0, 15, 30, 60 and 120 minutes in the process of dye removal. In all steps of the experiment, dye solutions with different initial concentrations of 100, 200, and 300 mg/L and the adsorbent dosage of 10 g/L were constantly used. Finally, pH of 3 and exposure time of 30 minutes were found as the optimum condition. The maximum dye removal (97%) was achieved at 100 mg/L of dye solution. Adsorbent specifications follow Langmuir and Freunlich isotherm model.

Key words: Sewage sludge, Dye Removal, Adsorbent; Isotherm models, Acid Blue25

INTRODUCTION

The increasing use of dyes in different industries has caused the discharge of toxic wastewater into the water resources resulting in serious environmental pollution (Aksu *et al.*, 2008; Tsai *et al.*, 2001; Ben Ticha *et al.*, 2013;Yamada *et al.*, 2013; Zaidan *et al.*, 2014; Varank *et al.*, 2014). Dyes are one of the important categories of aquatic pollutants present in the effluents of various industries such as textile, paper and pulp, printing, iron-steel, coke, petroleum, pesticide, paint solvent, pharmaceutics and wood preserving chemicals (Crini and Badot, 2008). The textile industries consume approximately 60% of total dyes produced to paint various fabrics, but around 10-15% of dyes used for this purpose come out through the effluent(Zollinger, 2004). Removal of dyes from the industrial effluents is a major environmental problem because most of these dyes are carcinogenic, mutagenic and teratogenic for human and other organisms (Cheng *et al.*,; Tsuby *et al.*, 2007; Golka *et al.*, 2004; Chung, 2005; Beaudoin and Pickering, 1960). Therefore, the removal of dyes from water and wastewater before mixing up with natural water resources is of special importance.

Conventional methods such as anaerobic treatment, trickling filters, floatation chemical coagulation, electrochemical coagulation, membrane separation, advanced oxidation process, photo degradation and adsorption are used for dye removal from water and wastewater (Delee *et al.*, 1998; Mondal, 2008; Rai *et al.*, 2005; Robinson *et al.*, 2001; Vanderzee and Villaverde, 2005).The production of adsorbent from various organic materials such as excess sludge produced in municipal wastewater treatment plants, industrial sludge, bone ash, coconut pith, pistachio crust, coconut crust, peach stone, olive pomace, bamboo, pine cone, etc have been investigated in various studies (Rita, 2012; Ladhe *et al.*, 2011; Kadirvelu *et al.*, 2009; Sivakumar and Palanisamy, 2009; Amina and Badia, 2008; Banat, 2007; Hameed *et al.*, 2007; Reddy *et al.*, 2006; Namasivayam and Kavitha, 2002; Ramakrishna, 1996). Adsorption using activated carbon is one of the most suitable techniques for

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removal of dyes from water and wastewater. Since it sometimes not economical due to the expensive cost, production adsorbents specially of activated carbon from sludge has been suggested by some researchers (Raddy *et al.*, 2006; Bardsal and Goyal, 2005).

This study aims to investigate the preparation of an adsorbent from sewage sludge to be used for the adsorption of one of industrial pollutants, namely Acid Blue 25 (L.A).

MATERIALS & METHODS

A digital laboratory scale with the accuracy of 0.0001 g (Sartorius Model) was used for weighting materials. Materials were dried in an electrical oven (Memmert model, Germany). pH of solutions were measured by a digital pH meter (CRISON Model, Spain). Dye solutions were mixed by an electrical shaker (Heidolph, Model: UNIMAX 2010). An electric furnace was used for carbonization and activation (Exciton Model). The dye concentration was measured by spectrophotometer (PG instruments Ltd T80+).

Required sludge for production of the adsorbent was collected from a municipal wastewater treatment plant located in Shahrak-e-Gharb district of Tehran, Iran. Acid Blue 25 (L.A.) is an acidic dye ($C_2OH_{13}N_2NaO_5S$) which was prepared from Alvan-Sabet Company and used as an industrial sludge in this study. It should be pointed out that the aforesaid dye is highly used in textile industries. In order to produce adsorbent, sludge was initially dried in an oven at 105°C for 24 hours. Then dried sludge was washed with HCl (98%) for two hours. To obtain pH of approximately neutral the sludge was washed by distilled water. Then, samples were carbonated and physically activated in a nitrogen-based furnace at 600C° for two hours. Carbonated and activated samples were crushed and powdered in a mortar and preserved in a sealed glass container to prevent moisture absorption. Molecular Structure of the adsorbent was determined through X-Ray Diffraction (XRD) technique.

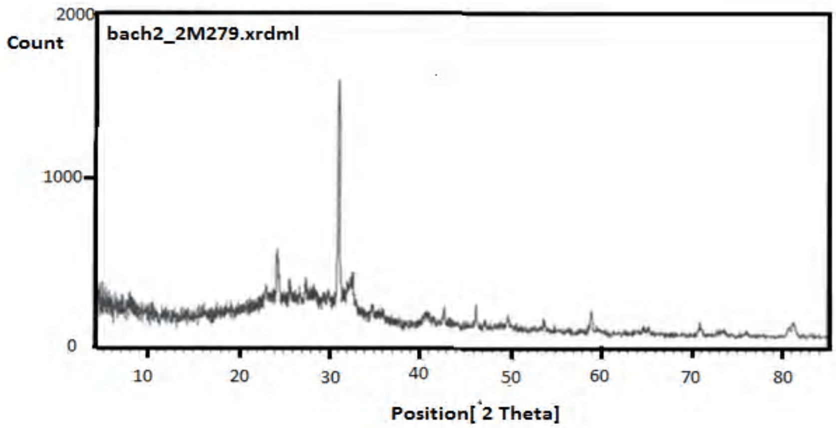
To provide the related calibration curve, the dye solutions prepared with 10, 25, 50, 75, and 100 mg/L concentration of the adsorbent to provide the related calibration curve. Then, the amount of dye-induced light absorption at 600 nm was measured by spectrophotometer. The calibration curve is used to obtain the concentration of unknown samples in comparison with the values obtained by spectrophotometer. In next step, dye solutions with different initial concentrations of 100, 200, and 300 mg/L and the adsorbent dosage of 10 g/L were used. Before adding the adsorbent, pH of solutions were fixed at 3, 5, 7, 9 and 11 by H_2SO_4 and NaOH. The solutions were placed on the shaker and stirred at 200 rpm for 0, 15, 30, 60, and 120 minutes. Finally, samples were filtered and their adsorption rates were measured by the spectrophotometer.

RESULTS & DISCUSSION

Determination of the components existing in the structure of an adsorbent is one of the important steps in the adsorption process. Results obtained from evaluation of adsorbent's structure revealed that the dominant compound of the adsorbent produced in this study is SiO_2. Fig. 1 shows XRD curve of the adsorbent. This molecular structure indicates that the adsorbent can be used for the removal of environmental pollutants due to the presence of Silica (a natural adsorbent). Furthermore, due to the presence of Hydroxyl-possessed compounds in the structure of the adsorbent, the ambient pH can play a significant role in the amount of pollutants adsorbed.

Results of the influence of pH and optimum exposure time on dye adsorption have been illustrated in Figs 2, 3, and 4. For dye concentrations of 100, 200 and 300 mg/L, maximum removal efficiencies obtained 97%, 85.75%, and 61.86%, respectively all at the same conditions (pH of 3 and exposure time of 30 minutes). The amount of dye adsorption increases with the decrease of pH so that maximum amount of adsorption obtained at the pH of 3. This result is in agreement with the results obtained by some other researchers who have carried out the same investigations (Sadri Moghaddam *et al.*, 2011; Sadri Moghaddam *et al.*, 2010a; Sadri Moghaddam *et al.*, 2010b; Mittal *et al.*, 2009a; Mittal *et al.*, 2009b; Hameed *et al.*, 2007). These researchers have stated that increase of pH leads to increase in OH^- and change in the adsorbent's surface charge. Consequently, dye adsorption decreases. Increase of exposure time leads to higher dye removal efficiency but the efficiency has not significantly changed after 30 minutes. Thus, this duration of time has been selected as the optimum exposure time. The fact that researchers have reported various exposure time for obtaining the highest dye removal efficiency, could be attributed to molecular structures differences of the applied adsorbents and the selected dye. The percentage of dye removal has decreased with the increase of concentration of dye solution from 100 to 300 mg/L. The same results have reported by Sadri Moghaddam et al. (2010a); Sadri Moghaddam et al. (2010b); Amin (2009); Sujatha et al. (2008a) and Sujatha, et al. (2008b).

In this study, using Langmuir and Freundlich adsorption isotherm models (equations 1 and 2) coefficient and equations have been derived and shown in Table 1. Where R^2=0.93 values of "a" and "b"



Peak list
01-085-0797;Si O2; Quartz
00-008-0048; K (Al , Fe) Si2 O8; Orthoclase
00-012-0204;Nax (Al, Mg)2 Si4O10 (O H)2 z H2 O; Montmorillonite
00-009-0466; Na Al Si3 O8; Albite, ordered
01-074-0345; K Al2(Si3 Al) O10 (O H)2; Moscovite

Fig.1. XRD curve of the adsorbent

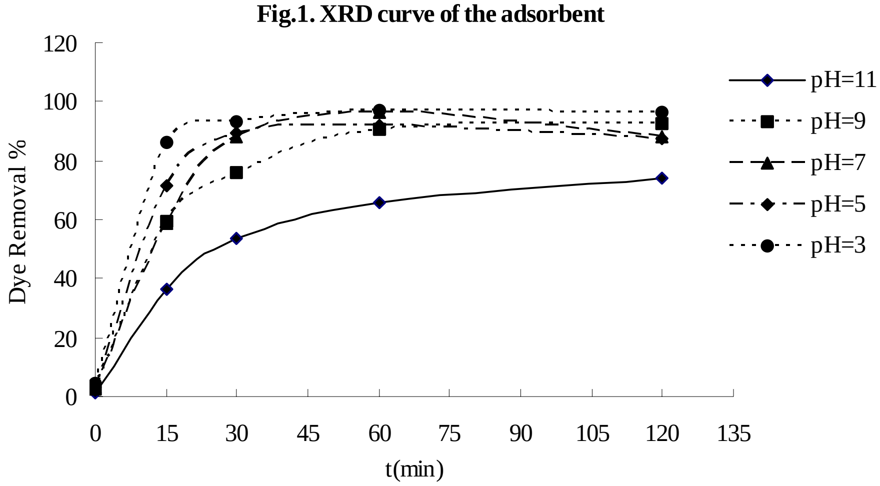


Fig. 2. Effect of pH and exposure time on dye removal (Adsorbent dosage=10 g/L, solution concentration =100 mg/L)

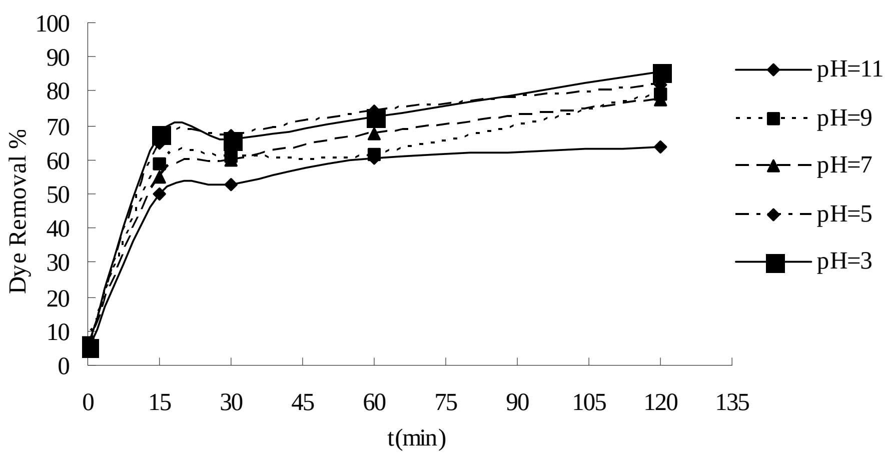


Fig. 3. Effect of pH and exposure time on dye removal (Adsorbent dosage=10 g/L, solution concentration =200 mg/L)

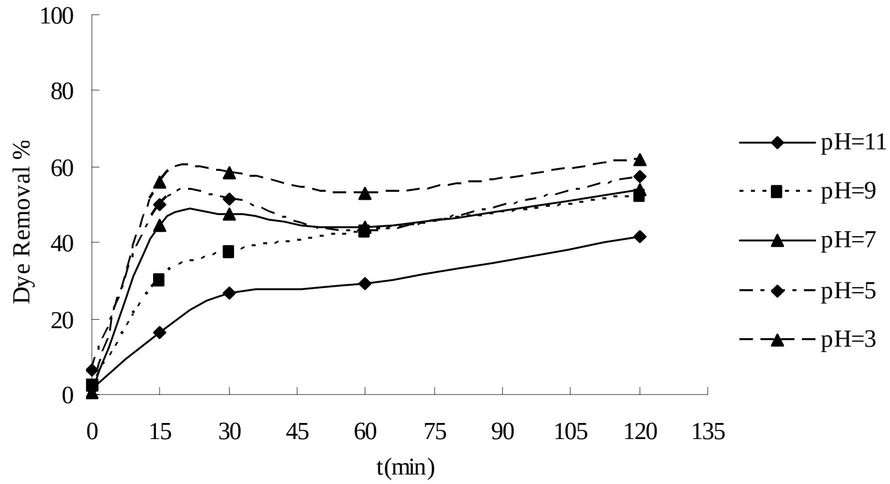


Fig. 4. Effect of pH and exposure time on dye removal (Adsorbent dosage=10 g/L, solution concentration =300 mg/L)

in Langmuir isotherm are equal to 0.42 and 14.94, respectively. Values of "K" and "n" in Freundlich isotherm where R^2=0.96, are 3.82 and 4.94, respectively.

$x/m=abc/1+ac$ (Benefield, 1982) (1)

x= amount of material absorbed (mg or g)

m=weight of adsorbent(mg or g)

c=concentration of material remaining in solution after adsorption is complete (mg/L)

a and b=constant

$x/m=KC^{1/n}$ (Benefield, 1982) (2)

x=amountof solute adsorbed (mg or g)

m=weight of adsorbent (mg or g)

C=concentration of solute remaining in solution after adsorption is complete (mg/L)

K and n= constant that must be evaluated for each solute and temperature

Comparison of the produced adsorbent isotherm model with Langmuir and Freunlich models (0.93 and 0.95) shows that produced adsorbent may follow both of the above mentioned isotherm models (Figs 5 and 6). However, for more accurate results, detailed experiments on dye concentration, amount of the adsorbent, exposure time and etc are recommended.

Table 1. Adsorption isotherms coefficients

Freundlich isotherm			Langmuir isotherm		
K	n	R^2	b	a	R^2
4.94	3.82	0.96	14.94	0.42	0.93

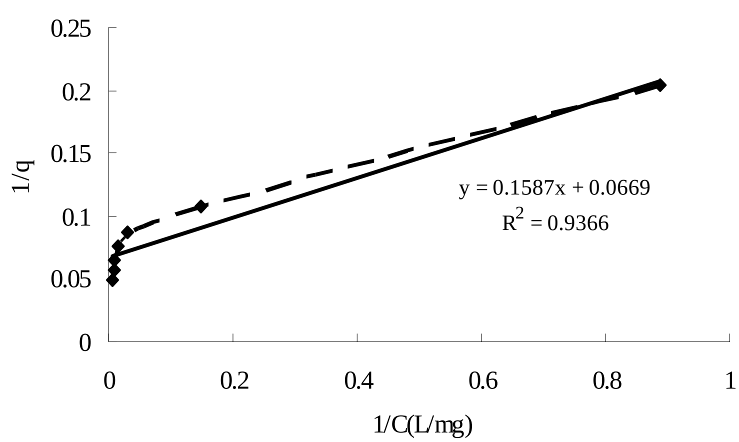


Fig. 5. Langmuir isotherm (pH=3; exposure time = 30 minutes; adsorbent dosage = 10 g/L)

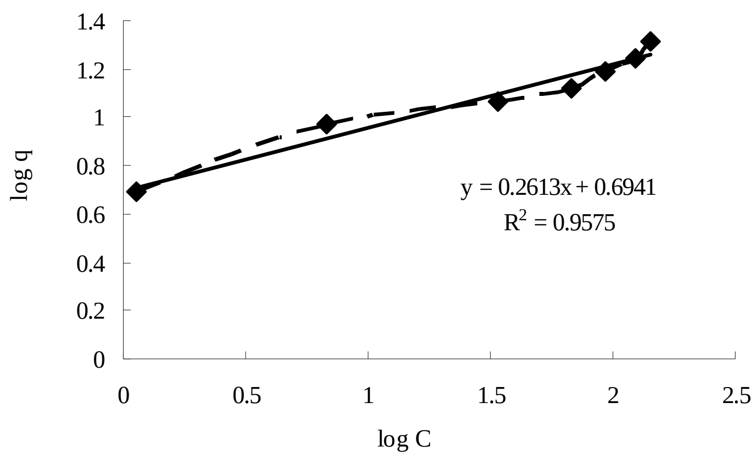


Fig. 6. Freundlich isotherm (pH=3; exposure time = 30 minutes; adsorbent dosage = 10 g/L)

CONCLUSION

In this study sewage sludge was used to produce an adsorbent for the removal of Acid Blue 25 dye. The results of XRD showed that the dominant compound of the produced adsorbent is SiO_2. The amount of dye adsorption increased with the decrease of pH so that maximum amount of adsorption obtained at the pH of 3. Increase of exposure time lead to higher dye removal efficiency but the efficiency did not significantly change after 30 minutes. Therefore, pH of 3 and exposure time of 30 minutes were found as the optimum condition. The percentage of dye removal decreased with the increase of concentration of dye solution from 100 to 300 mg/L. Moreover, the results indicated that produced adsorbent may follow both of the above mentioned isotherm models. However, for more accurate results, detailed experiments on dye concentration, amount of the adsorbent, exposure time and etc are recommended. Results of the present study showed that adsorption process using the adsorbent produced from sewage sludge could be considered as a practical and efficient method for dye removal.

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