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Treatment of Textile Waste Water by Adsorption Using Granular Activated Carbon Along with Isotherm Modeling and Kinetics

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TREATMENT OF TEXTILE WASTE WATER BY ADSORPTION USING GRANULAR ACTIVATED CARBON ALONG WITH ISOTHERM MODELING AND KINETICS

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Abstract

Textile industries are one of the major polluters in recent times. These industries produce huge amount of waste water which are highly in need of proper treatment. The project focuses on the treatment of textile waste and dyes with Granular Activated Carbon (GAC) by adsorption process. After treatment with 4g GAC/L wastewater, the Chemical Oxygen Demand (COD) of sample waste water reduced almost 65%. A huge change was also found in the physical appearance of the sample water. After treatment almost 86% color was gone. The pH also increased by about 5%. Four isotherm models-Linear, Langmuir, Freundlich and Temkin were studied. Linear isotherm showed the best fitting with the equilibrium data in terms of the coefficient of determination (R^2). In the kinetics study, pseudo first-order, pseudo second-order, Elovich's equation and intra particle diffusion kinetic models were studied. Elovich's equation well fit the experimental data.

Keywords: Adsorption-isotherm, Adsorption-kinetics, Granular activated carbon, Textile wastewater, Dyestuffs.

1. Introduction

Granular activated carbon is used for the treatment of industrial waste water and many other water supplies. It provides a versatile technology suitable for removing a broad range of both organic and inorganic pollutants. These pollutants are generally of concern because of their toxicity to human health and existence of aquatic animals. [1] Some contaminants have a special risk due to their high frequency of occurrence in surface waters, among which non-biodegradable organic compounds (COD), adsorbable organic halogens (AOX), toxicity, color compounds and dyestuffs, inhibitory compounds for biological treatment systems, aromatic compound including phenol and bis-phenol A (BPA), chlorinated or halogenated organic compounds and pesticides are common. [4] Today we have our modern and civilized world. But to get to this point, the things that have grown along with population are the industries. Textile industries are second largest industries globally. According to a

report of World Bank, almost 20% of global industrial water pollution comes from the treatment and dyeing of textiles [2]. As an economically growing country, the textile industries are spreading its wings in Bangladesh as well. Almost 80% of national exports in Bangladesh are based on Textile Mills. So it is one of the most important industries in Bangladesh. But every blessing has a dark side and so does these industries. The problem is that these textile industries produce waste water which causes environmental problems, mostly pollution of water resources of the country. The effluent waste water contains not only dissolved dye, but also poisonous metallic ions, aromatic amines, ammonia, alkaline salts and large amount of pigments. Some of these chemicals cannot be filtered or removed. These particles, if comes in contact with living beings may cause fatal diseases and destruction of the natural system. So the treatment of the textile waste water is very important.

GAC is generally made from organic materials with high carbon contents such as wood, lignite and coal. The basic characteristic that differentiates GAC to PAC is its particle size. Typically, GAC has a diameter ranging between 1.2 to 1.6 mm and an apparent density ranging between 400.462 and 496.572 kg/m³, depending on the material used and manufacturing process. The bed density is about 10 percent less than the apparent density and is used to determine the amount of GAC required to fill a given size filter. The uniformity coefficient of GAC is quite large, typically about 1.9, to promote stratification after backwashing and minimize desorption and premature breakthrough that can result from mixing activated carbon particles with adsorbed compounds with activated carbon particles with smaller amounts of adsorbed compounds. Iodine and molasses numbers are typically used to characterize GAC. These numbers describe the quantity of small and large pore volumes in a sample of GAC. A minimum iodine number of 500 is specified for activated carbon by AWWA standards. [3]

Activated carbon is one of the most effective media for removing a wide range of contaminants from industrial and municipal waste waters, landfill leachate and contaminated groundwater. As the world's most powerful adsorbent, it can cope with a wide range of contaminants. Different contaminants may be present in the same discharge and carbon may be used to treat the total flow, or it may be better utilized to remove specific contaminants as part of a multistage approach.

Once granular activated carbon is saturated, or the treatment objective is reached, it can be recycled, by thermal reactivation for reuse. Reactivation involves treating the spent carbon in a high temperature reactivation furnace to over 800°C. During this treatment process, the undesirable organics on the carbon are thermally destroyed. Recycling activated carbon by thermal reactivation meets the

environmental need to minimize waste, reducing CO₂ emissions and limiting the use of the world's resources. [4]

The project is mainly on the treatment of textile waste and dyes with coconut shell based Granular Activated Carbon (GAC) by adsorption process. Erlen Meyer flasks were used for conducting the adsorption process in a 24 hour run in batch process. Different amounts of GAC, from 0.09g to 1.5g were put in a fixed volume of 150 ml of sample waste water. Then the solution was filtered and the filtrate was collected and corresponding tests were conducted. After figuring the optimum amount of GAC, a sample was made with that amount and collected at different times of 24 hour run to find out the kinetics and isotherms. In this experiment, it is mainly focused to remove the color compound of dyes, COD and total dissolved solids. With further study and modification on this project it could be very effective in the treatment of textile wastes and to prevent the environmental pollution in Bangladesh and also the whole world.

2. Literature Review.

The process by which liquid or gaseous molecules are concentrated on a solid surface can be referred to as adsorption. In this case the solid surface is activated carbon. This is different from absorption process, where molecules are taken up by a liquid or gas. [4]

Activated carbon is commonly used to adsorb natural organic compounds, taste and odor compounds, and synthetic organic chemicals in water treatment. Adsorption is both the physical and chemical process of accumulating a substance at the interface between liquid and solids phases. Activated carbon is an effective adsorbent because it is a highly porous material and provides a large surface area to which contaminants may adsorb. The two main types of activated carbon used in water treatment applications are granular activated carbon (GAC) and powdered activated carbon (PAC).

However, commercially available activated carbon is expensive. In the last years, special emphasis on the preparation of activated carbons from several agricultural by-products has been given due to the growing interest in low cost activated carbons from renewable, copious, especially for application concerning treatment of wastewater. Researchers have studied the production of activated carbon from palm-tree cobs, plum kernels, cassava peel, bagasse, jute fiber, rice husks, olive stones, date pits, fruit stones and nutshells. The advantage of using agricultural by-products as raw materials for manufacturing activated carbon is that these raw materials are renewable and potentially less expensive to manufacture. [10]

The two most common options for locating a GAC treatment unit in water treatment plants are: (1) post-filtration adsorption, where the GAC unit is located after the conventional filtration process (post-filter contactors or adsorbers); and (2) filtration-adsorption, in which some or all of the filter media in a granular media filter is replaced with GAC. In post-filtration applications, the GAC contactor receives the highest quality water and, thus, has as its only objective the removal of dissolved organic compounds. Backwashing of these adsorbers is usually unnecessary, unless excessive biological growth occurs. This option provides the most flexibility for handling GAC and for designing specific adsorption conditions by providing longer contact times than filter-adsorbers.

In addition to dissolved organics removal, the filter-adsorber configuration uses the GAC for turbidity and solids removal, and biological stabilization. Existing rapid sand filters can frequently be retrofitted for filtration-adsorption by replacing all or a portion of the granular media with GAC. Retrofitting existing high rate granular media filters can significantly reduce capital costs since no additional filter boxes, underdrains and backwashing systems may be required. However, filter-adsorbers have

shorter filter run times and must be backwashed more frequently than post-filter adsorbers (filter-adsorber units are backwashed about as frequently as conventional high rate granular filters). In addition, filter-adsorbers may incur greater carbon losses because of increased backwashing and may cost more to operate because carbon usage is less effective.

Primary factors in determining the required GAC contactor volume are the (1) breakthrough, (2) empty bed contact time (EBCT), and (3) design flow rate. The breakthrough time is the time when the concentration of a contaminant in the effluent of the GAC unit exceeds the treatment requirement. As a rule of thumb, if the GAC effluent concentration is greater than the performance standard for over three consecutive days, the GAC is exhausted and must be replaced/regenerated. The EBCT is calculated as the empty bed volume divided by the flow rate through the carbon. Longer EBCTs can be achieved by increasing the bed volume or reducing the flow rate through the filter. The EBCT and the design flow rate define the amount of carbon to be contained in the adsorption units. A longer EBCT can delay breakthrough and reduce the GAC replacement/regeneration frequency. The carbon depth and adsorber volume can be determined once the optimum EBCT is established. Typical EBCTs for water treatment applications range between 5 to 25 minutes.

The carbon usage rate (CUR) determines the rate at which carbon will be exhausted and how often carbon must be replaced/regenerated. Carbon treatment effectiveness improves with increasing contact times. Deeper beds will increase the percentage of carbon that is exhausted at breakthrough. The optimum bed depth and volume are typically selected after carefully evaluating capital and operating costs associated with reactivation frequency and contactor construction costs.

Depending on the economics, facilities may have on-site or off-site regeneration systems or may waste spent carbon and replace it with new. Spent GAC must be disposed of recognizing that contaminants can be desorbed, which can potentially result in leaching of contaminants from the spent GAC when exposed to percolating water, contaminating soils or groundwater. Due to contamination concerns, spent GAC regeneration is typically favored over disposal. The three most common GAC regeneration methods are steam, thermal and chemical; of which thermal regeneration is the most common method used. Available thermal regeneration technologies used to remove adsorbed organics from activated carbon include: (1) electric infrared ovens, (2) fluidized bed furnaces, (3) multiple hearth furnaces, and (4) rotary kilns. [3]

3. Research Methods

3.1 Materials

The adsorbent was coconut shell based granular activated carbon which is used commercially. The activated carbon crystals were of 8x16 mesh number, pH value 7, specific surface area 1060 m²/g, hardness 95% and 0.51 g/cm³ bulk density. Textile dyeing wastewater was collected from a local industry in Gazipur, Dhaka, Bangladesh.

3.2 Experimental

Equilibrium adsorption isotherm study was carried out using bench top shaker. To run the experiment in a batch operation, different amounts of GAC weighing 0.09 g to 1.5 g were measured using a digital electronic weighing machine. These measured amounts of GAC were then placed into 250 ml Erlen Meyer flasks. Sample wastewater was added into each flask and the volume was made 150 ml. Then the flasks were covered with aluminum foil paper to prevent contamination. The flasks were then put onto a shaker for 24 hours. A blank experiment (without addition of GAC) was

also carried out to measure the degradation of organics without GAC. The experiment was carried out at constant speed of 250 rpm. After the experiment, the samples with GAC were filtered using a vacuum filtration unit. Whatman® 42 quantitative filter paper (diameter 9.0 cm, 2.5 µm pore, Sigma Aldrich) was used to filter the treated wastewater for removing the GAC crystals. The filtrate was collected to measure COD, color and other the physical and chemical properties.

COD was measured according to standard methods (American Public Health Association, 2005). To measure the COD, 4 ml of COD reagents and 3 ml of dilute Sulfuric Acid solutions were added to a 2 mL sample. These samples were placed onto a HACH DRB 200 COD reactor and digested at 150° C for 2 hours. The COD vials were taken out from the reactor and cooled to room temperature. The absorbance was measured by a HACH Spectrophotometer (DR 3900 Benchtop VIS Spectrophotometer with RFID Technology). The color of raw and treated wastewater was measured using standard methods (APHA 2005).

3.3 Adsorption Isotherms

Equilibrium study on adsorption provides information on the capacity of the adsorbent. An adsorption isotherm is characterized by certain constant values, which express the surface properties and affinity of the adsorbent and can also be used to compare the adsorptive capacities of the adsorbent for different pollutants. The experimental data were fitted using different equilibrium models (Linear, Langmuir, Freundlich and Temkin) and kinetic models (pseudo first-order, pseudo second order, Elovich's equation and intra-Particle diffusion kinetic models). The amount of organics adsorbed per unit adsorbent mass was calculated as follows [5]:

$$q_e = \frac{(C_i - C_e)V}{m}$$

Where, C_i is the initial concentration of COD in wastewater, mg/L; C_e is the equilibrium concentration of COD in wastewater after treatment, mg/L; V is the volume of wastewater, L; and m is the mass of GAC used, g. The removal efficiency of organics can be calculated from the equation below:

$$\% \text{ removal} = \frac{(C_i - C_e) \cdot 100}{C_i}$$

3.3.1 Freundlich isotherm

The Freundlich adsorption isotherm assumes that adsorption occurs on a heterogeneous surface through a multilayer adsorption mechanism and that the adsorbed amount increases with the concentration according to the following equation [6]:

$$q_e = K_f C_e^{1/n}$$

Where q_e is the amount adsorbed at equilibrium in terms of (mg organics adsorbed/g PAC), C_e is the equilibrium COD concentration, K_f (L/g) is the Freundlich constant and $(1/n)$ is related to the strength of adsorption driving force and the degree of nonlinearity between solution concentration and adsorption. The plot of $\ln q_e$ versus $\ln C_e$ is employed to generate K_f value from the intercept and n value from the slope. The linear form of Freundlich equation is:

$$\ln q_e = \frac{1}{n} \ln C_e + \ln K_f$$

3.3.2 Langmuir isotherm

The Langmuir model considers several assumptions: the adsorption is localized, all the active sites on the surface have similar energies, none interaction between adsorbed molecules exists, and the limiting reaction step is the surface reaction as in the heterogeneous catalytic reaction. The Langmuir equation can be written as [6]:

$$q_e = \frac{q_m K_L C_e}{1 + K_L C_e}$$

where C_e and q_e have the same meaning as in the Freundlich isotherm. q_m is the maximum uptake per unit mass of carbon (mg/g), and K_L is the Langmuir constant related to the adsorption energy (L/mg). The linear form of Langmuir equation is:

$$\frac{1}{q_e} = \frac{1}{q_m K_L C_e} + \frac{1}{q_e}$$

3.3.3 Temkin isotherm

Temkin and Pyzhev suggested that, because of the existence of adsorbate–adsorbate interactions, the heat of adsorption should decrease linearly with the surface coverage. The linear form of Temkin isotherm model is given by the equation [6]:

$$q_e = \frac{RT}{b} \ln K_T + \frac{RT}{b} \ln C_e$$

$$\text{Or, } q_e = B \ln A + B \ln C_e$$

Where, b is the Temkin constant related to the heat of sorption, J/mol and K_T is the Temkin isotherm constant, L/g, R is the universal gas constant, J/mol/K and T is the liquid temperature, K.

3.3.4 Linear Isotherm

The linear isotherm model can be expressed by the following equation:

$$q_e = k \cdot C_e$$

where q_e is amount of solute adsorbed per unit weight of solid at equilibrium in g/g or mg/g and C_e is the equilibrium concentration of solute remaining in the solution, when amount adsorbed equals q_e .

3.4 Adsorption Kinetics

3.4.1 Pseudo first-order model or Lagergren's equation

The pseudo first-order equation of Lagergren is one of the most widely used for the adsorption of solute from a liquid solution. The linear form can be written as [6]:

$$\ln(q_e - q_t) = \ln q_e - k_1 t$$

where k is the kinetic constant of pseudo first-order adsorption (h⁻¹), and q_e and q_t (mg/g AC) represent the amounts of adsorbed amount at equilibrium and at time t (h), respectively.

3.4.2 Pseudo second order

A pseudo second-order equation based on equilibrium adsorption can be expressed as [6]:

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e}$$

where k_2 (g/mg h) is the rates constant of second-order adsorption. If second-order kinetics is applicable, the plot of t/q versus t should show a linear relationship.

3.4.3 Intra-particle diffusion model

The sorption kinetics may alternatively be described from a mechanistic point of view. The overall adsorption process may indeed be controlled either by one or more steps, e.g. film or external diffusion, pore diffusion, surface diffusion and adsorption on the pore surface, or a combination of more than one step. The possibility of intra-particle diffusion was explored by using the intra-particle diffusion model, according to which the amount adsorbed at time t , q_t , reads [6]:

$$q_t = k_{id} \cdot t^{1/2} + \Theta$$

Where, K_{id} is the intraparticle diffusion rate constant, mg/g; and Θ is the intercept, mg/g.

3.4.4 Elovich's equation

Elovich's equation describes activated adsorption. It was established through the work of Zeldowitsch dealing with the adsorption of carbon monoxide on manganese dioxide Elovich's equation has also been used for describing successfully the adsorption of pollutants from aqueous solutions in recent years and can be expressed as follows [6]:

$$q_t = \frac{1}{b} \ln(ab) + \frac{1}{b} \ln(t)$$

where a is the initial adsorption rate (mg/(g h)), and the parameter $1/b$ (mg/g) is related to the number of sites available for adsorption. If this equation applies, it should lead to a straight line by plotting q_t as a function of $\ln t$.

4. Analysis & Results

The wastewater was treated in a batch study using different GAC doses for the study of different isotherm models. However, for the

study of adsorption kinetics model, a separate batch study with optimum GAC dose (4g GAC/L wastewater) for the treatment of wastewater was selected and sampling was performed at different time interval.

4.1. Equilibrium Adsorption Performance

The concentrations of COD both raw and treated wastewater were measured and Figure 1 shows the removed concentration in mg/L with the GAC dose in g/L. The COD removal efficiencies enhanced with increasing in GAC dosage; whilst decreasing in the equilibrium adsorbed amount per g of GAC. For example, 0.6 g/L of GAC dosage removed 41% COD, which increased to 68% after addition of 4 g/L GAC dose. Further increase of GAC dose from 4 g/L to 10 g/L didn't increase any COD removal efficiencies considerably.

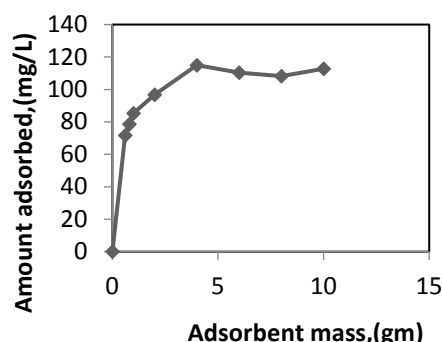


Fig 1: Equilibrium adsorption curve for determining the optimum dose of GAC

However, the amount of COD adsorbed on GAC increased from 60 mg/L for 0.5 g GAC/L wastewater to 120 mg/L for 4 g GAC/L wastewater as shown in the Figure 1. This could be explained that at higher GAC dosage, the ratio of the initial concentration of organic matters (COD) to the GAC available site is low and subsequently the fraction of sorption is independent of the initial concentration. Thus, a lower removal of COD per g of GAC (but higher COD removal efficiency) was observed. On the other hand, at the lower GAC dosage, the ratios of the initial concentration of organic matters (COD) to

the GAC available site is high and thus increase the adsorption capacity per g of GAC. The available adsorption sites became fewer compared to the amount of organic matters and hence resulted in the lower COD removal efficiency (Xing et al., 2008).

4.2 Isotherm evaluation and study

Description of adsorption equilibrium by an appropriate isotherm is the most important step to design an adsorption system as it reflects the capacity or affinity of an adsorbent for a particular adsorbate. Adsorption of COD by GAC from textile dyeing wastewater was

modeled using Linear, Freundlich, Langmuir and Temkin isotherms and Figure 3 shows those isotherms. The quality of the fit of isotherms was assessed using the correlation coefficient. All the parameters of the four different studied isotherms and the values of their corresponding correlation coefficients (R^2) for all isotherms from the isotherm plots are listed in Table 1. The q_m is attributable to the saturation amount of organic matters adsorbed in the Langmuir isotherm. Based on the results listed in Table 1, best isotherm models fitted for COD removal are determined in the order: Linear > Temkin > Freundlich > Langmuir.

Table 1: Parameters and correlation coefficients of Linear, Langmuir, Temkin and Freundlich Isotherms.

Linear		Langmuir			Freundlich			Temkin		
R^2	K (L/g)	K_L (L/mg)	q_m (mg/g)	R^2	n	K_f (mg/g)	R^2	K_t (L/mg)	B	R^2
0.951	2.45	0.0091	11.76	0.638	0.243	1.819	0.846	$e^{-4.08}$	196.3	0.933

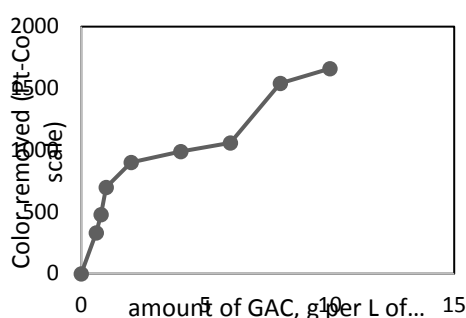


Figure 2: Removed Color on Pt-Co scale with different doses of GAC

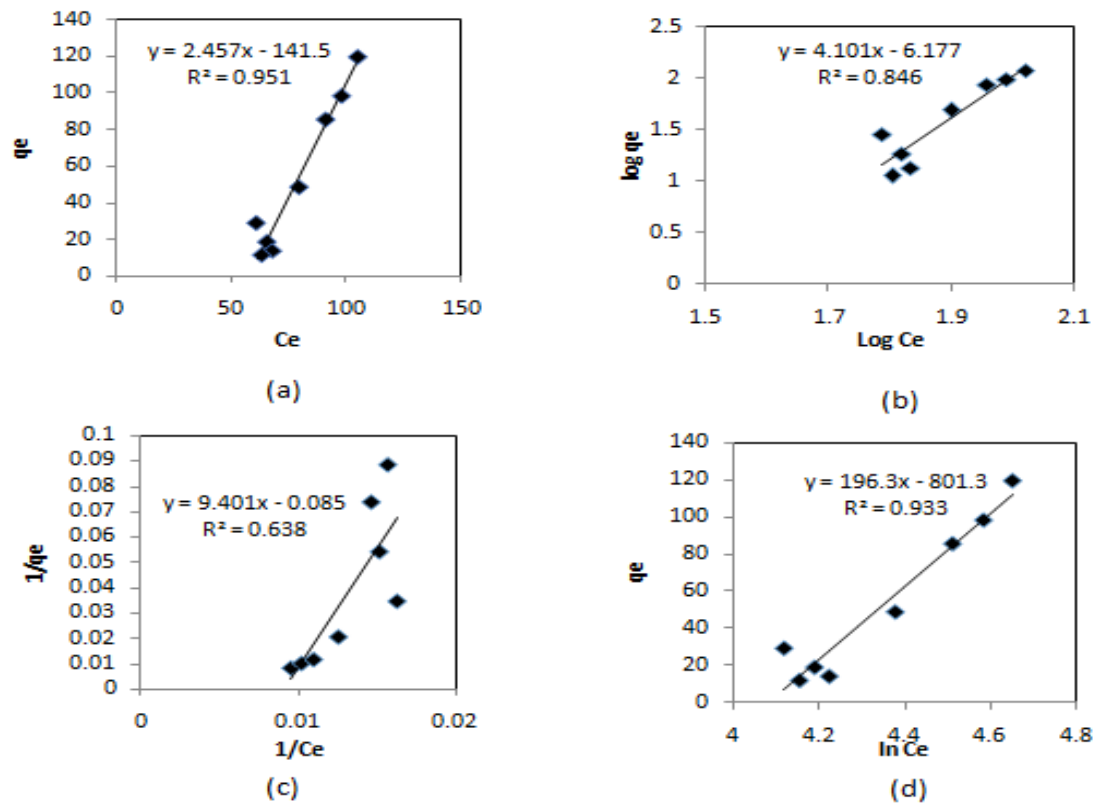


Figure 3: (a) Linear (b) Freundlich (c) Langmuir and (d) Temkin isotherms for adsorption Modeling study.

4.1. Kinetic study

As in the Figure 1, we observed that the 4 g GAC/L wastewater was optimum GAC dose for COD removal. Thus the 4 g/L GAC

dose was selected as optimum GAC dose for adsorption kinetics study.

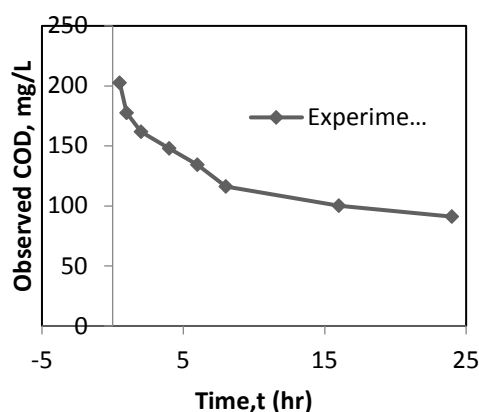


Figure 4: COD concentration change during experiment with time.

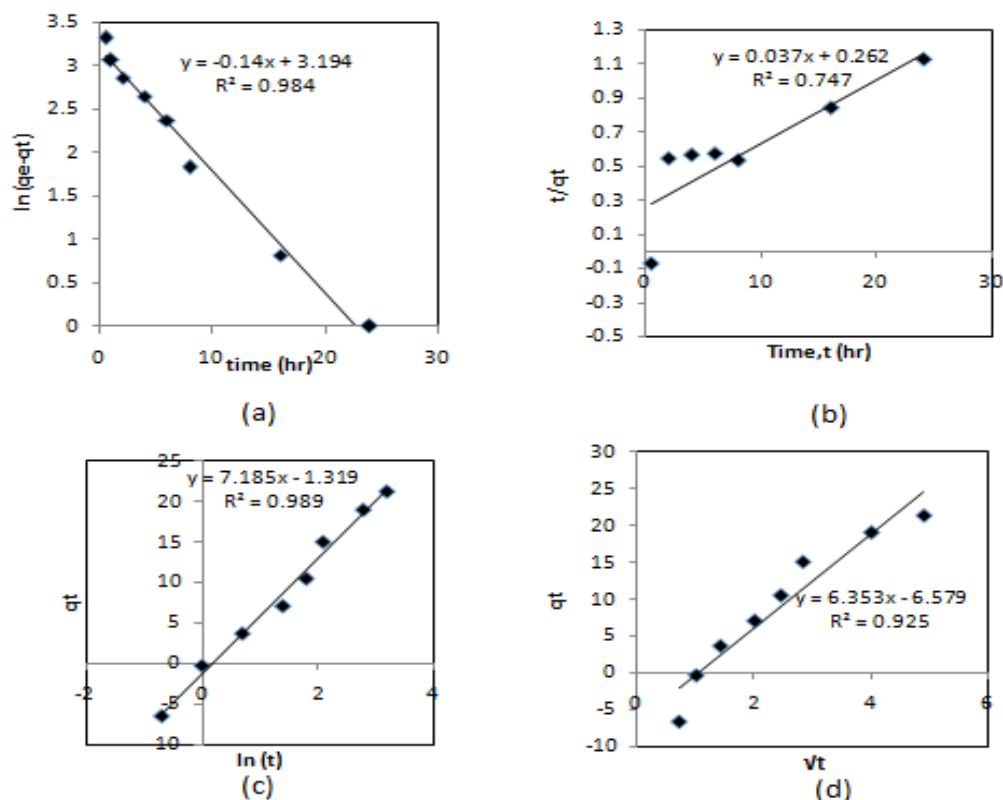


Figure 5: Application of the (a) Pseudo first-order (b) Pseudo second-order (c) Elovich's equation (d) Intra-Particle diffusion for the experimental data.

Figure 4 shows the graphical representation of adsorption characteristics of measured COD of the sample as a function of time. It was observed that initially the COD removal was fast and within 2 hrs COD reduced by almost 65%. Then the rate became slow because of less concentration gradient of COD between bulk liquid and onto the GAC surface and it reached equilibrium within 6 hrs. In order to analyze the adsorption kinetics, correlations between adsorbed amounts and time were sought for, through the testing of different

mathematical expressions corresponding to various models, namely: pseudo first-order, pseudo second-order reaction and then Intra-particle diffusion and Elovich's equation. Figure 5 represents these models. Table 2 represents the kinetic parameters from all the studied models. . From the Table it is observed that Elovich's equation fitted best with the experimental data as the R^2 value is 0.989 here. So the adsorption of GAC in dyeing textile wastewater follows Elovich's equation.

Table 2: Obtained kinetic parameter from the adsorption process

Pseudo first-order			Pseudo second-order			Elovich's equation			Intra-Particle diffusion		
k (h ⁻¹)	q _e (mg/g)	R ²	k' (g/(mg·h))	q _e (mg/g)	R ²	a (mg/(g·h))	1/b (mg/g)	R ²	K _{id} (mg/(g·h ^{1/2}))	Θ (mg/g)	R ²
0.140	24.38	0.984	0.005	27.02	0.747	0.167	7.194	0.989	6.353	6.579	0.925

5. Discussion & Conclusion

This study demonstrated that GAC is an excellent adsorbent for removal of waste and color from textile waste. Within 24 h, 4 g/L GAC could remove 78% of color. Adsorption behaviour is described by a Linear isotherm model among four adsorption isotherm equilibrium models - Linear, Freundlich, Temkin, and Langmuir. The classification of the kinetic models according to the simulation of the adsorption study is: Elovich's equation > Intra-particle-diffusion > Pseudo-first order > Pseudo-second order Kinetic model. So the experimental data follows Elovich's equation model which is actually a chemisorption process for the adsorption of GAC over time.

GAC is already used by water treatment plants on either a full time basis or as needed for taste and odor control or removal of organic chemicals. Application of GAC generates additional sludge which is not likely to be classified as a hazardous waste. However, the results from this experiment suggested that the GAC is a promising adsorbent for the removal of organics from textile dyeing wastewater. Thus, further study of GAC in combination with coagulation-flocculation and biological processes is necessary to achieve effective utilization of GAC in the wastewater treatment. This modification on

this project could be more effective in the treatment of textile wastes and to prevent the environmental pollution in Bangladesh.

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