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Studies on Removal of Methylene Blue Color Using Waste Tea Power as an Adsorbent

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Abstract

The studies of adsorption were carried out in batch scale to estimate the different adsorption parameters concerning the efficiencies of dye removal from aqueous solutions. The data for contact time obtained from the adsorption of methylene blue on the waste material such as Waste Tea Powder (WTP) show that a contact time of 25 min and there was no significant change in concentration of the dye with further increase in contact time. The strategy of pH versus percentage adsorption shows the significant adsorption takes place with WTP for dye at 6 only. The uptake equilibrium was increased and adsorption percentage was decreased with increasing the initial dye concentration for the adsorbent. The percentage adsorption of dye increases with increasing the adsorbent dosage of WTP. The percentage adsorption of dye decreases with increasing WTP particle size. The percentage adsorption and dye uptake of dye increases with increasing the temperature of solution with WTP. The Langmuir isotherm for both the adsorbents proved to be the best adjustment of the experimental data than the Freundlich Isotherm.

Keywords: Waste Tea Powder, Adsorption, Langmuir Isotherm, Freundlich Isotherm,MBD.

Introduction

Dyes and pigments have been used in many industries for coloration purpose. Textile industry is one of the prominent polluters releasing high concentrated effluent into the surrounding environment. Dyes and pigments have been used in many industries for coloration purpose. Textile industry is one of the prominent polluters releasing high concentrated effluent into the surrounding environment. Over $7x10^5$ tones and approximately 10,000 different types of dye and pigment are produced worldwide annually and the volume is steadily increasing. Dyes contain carcinogenic materials which can pose serious hazards to aquatic life and end users of the water. Dyes and pigments are widely used, mostly in the textiles, paper, plastics, leather, food and cosmetic industry to color products. The release of colored wastewater from these industries may present an eco-toxic hazard. Dyes are an important class of pollutants, and can even be identified by the human eye. Disposal of dyes in precious water resources must be avoided, however, and for that various treatment techniques like precipitation, ion exchange, chemical oxidation, and Adsorption have been used for the removal of toxic pollutant from, wastewater.

Methylene blue is a heterocyclic aromatic chemical compound with the molecular formula $C_{16}H_{18}N_3SCl$. It has many uses in a range of different fields, such as biology and chemistry. At room temperature it appears as a solid, odorless, dark green powder. So that it yields a blue solution when dissolved in water. The hydrated form has 3 molecules of water per molecule of methylene blue. Methylene blue was chosen for this study because of its known strong Adsorption on to solids. Methylene Blue is the most commonly used material for dying cotton, wood, and silk with molecular weight 373.9 corresponds to methylene blue hydro chlorine with three groups of water.

The International Nonproprietary Name (INN) of methylene blue is methylthioninium chloride.

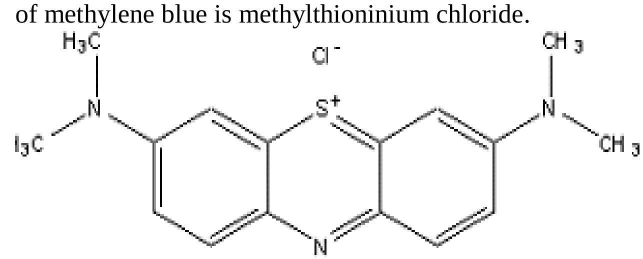

Fig 1.1 Methylene Blue structure

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Behavior of Methylene Blue in solution

Methylene blue is widely used as a redox indicator in analytical chemistry. Solutions of this substance are blue when in an oxidizing environment, but will turn colorless if exposed to a reducing agent. The redox properties can be seen in a classical demonstration of chemical kinetics in general chemistry, the "blue bottle" experiment. Typically, a solution is made of dextrose, methylene blue, and sodium hydroxide. Upon shaking the bottle, oxygen oxidizes methylene blue, and the solution turns blue. The dextrose will gradually reduce the methylene blue to its colorless, reduced form. Hence, when the dissolved oxygen is entirely consumed, the solution will turn green.

Sources of Methylene Blue and its availability

Methylene Blue starts its journey as an extremely valuable dye-a dye of such importance that it not only revolutionized the textile industry but also resulted in a patent challenge that changed intellectual property law.

Until the middle of 19th century, only natural dyestuffs were available for use by dyers. These offered a fairly limited color palette, and many stains faded easily upon washing and exposure to light. The synthetic dye industry was born in 1856, when an English Chemist, Sir William Henry Perkin, in inadvertently created the first aniline dye while attempting to make quinine from coal-tar derivatives. Perkin named his new dye aniline purple, but it soon became known as mauve after the French name for the flower with a similar color. Mauve was an instant triumph, and soon the European fashion Centre of London and Paris bustled with purple garments. In subsequent decades, a rainbow of other aniline dyestuffs were synthesized and made available to textile colorists. Germen companies almost all are these dyes, and the Centre of the textile industry quickly shifted away from Perkin’s England. Anilinfarbenfabrikation (AGFA), made a fortune from methylene blue, exporting it to textile manufacturers all over the world. Other dye companies soon found themselves unable to compete. AGFA’S product was so successful that competitors had to close factories lay of workers.

Methylene Blue is first prepared by Caro in 1876 as an aniline derived dye form textiles. However it is drug full of surprises that has made history as a histochemical stain, a Biological reagent and lead compound in the development of therapeutic agents for diseases ranging from microbial disease to dementia. The staining activity of Methylene Blue, developed by Paul Ehrlich in 1891, provided the foundation of modern chemotherapy. In the late 1919 and early 20th centuries it was used in humans to treat malaria, but then ceased to be used as an anti-malarial due to its inevitable side effects.

Adsorption is an efficient and economical method for removing dyes from industrial effluents. In this process, a substance (soluble dye) from the liquid phase (wastewater) is transferred to the surface of a solid, highly porous material (adsorbent), to which it binds physically or chemically. The adsorption technique is preferable to other wastewater treatment techniques in terms of efficiency, low cost, simplicity, ease of operation, and inactivity towards toxic substance. Moreover, the specific advantage of method is that the adsorbent can be chosen from a large variety of material. These adsorbent, used in batch or dynamic condition, can be naturally occurring material (wood, peat, coal, chitin and chitosan, biomass, clays,etc.), as well as industrial/agricultural wastes or byproducts (fly ash, red mud, blast furnace slag, dye hydroxide sludge, sawdust, bark, lignin, sunflower stalks, maize cob, rice husk, hazelnut shell, olive stones, seashell, etc.). The aim of this work is to evaluate the efficiency of using (Tea powder and coffee powder) as adsorbent for removal of methylene blue (MB) from an aqueous environment. The obtained result gives some insight with respect to utilization of (Tea powder and coffee powder) as an eco-friendly adsorbent in textile industry wastewater treatment. Thus, these results may be applied for predicting the adsorption mechanism, for characterization and optimization of the process, and equipment and process design.

Large quantities of dyes are utilized by the textile industry. Mainly due to their extensive application in the dyeing of cotton, about 20-30% of all of the dyestuffs used worldwide are reactive dyes. Biodegradation of this group of compounds is significantly slower than that of acid and direct dyes. The persistent nature of reactive dyes complicates wastewater treatment within the textile industry. Removal of dyestuffs from wastewater occurs by either biological methods or physicochemical methods (e.g., adsorption, oxidation-reduction, chemical coagulation, ozone treatment, and membrane filtration). Low cost adsorbents have gained attention over decades as a means of achieving very high removal efficiency to meet discharge standards. The WTP is a potentially useful material for the removal of dyes from industrial wastewater. It is suggested that the work is designed to increase the scale WTP application to cope with the removal of methylene blue component under industrial conditions should be initiated. The WTP has a

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substantially larger capacity to remove the reactive dye by Adsorption than all of the other components.

The performance analysis was carried out as function of various operating parameters such as contact time, initial concentration of dye, adsorbent dose, particle size and pH. Performance studies revealed that a very high percentage removal of color was achievable. Detailed data analysis indicated that the Adsorption of methylene blue followed Freundlich isotherm and Langmuir isotherm. Comparative study was also done on the Adsorption of methylene blue with other adsorbents on the basis of contact time.

Experimental Procedure

Preparation of adsorbent Waste Tea Powder-WTP

After collecting the WTP sample from nearby sources, it is cleaned by washing with water and dried in the atmosphere. After drying it is separated into various sizes by using BSS sieves.

Preparation of Methylene Blue solution

Stock solution of Methylene Blue concentration (1000 mg/L) was prepared by diluting 1 g of Methylene Blue powder in a 1000 ml of distilled water. Later it was diluted to get the test solutions of concentrations 0.02 to 0.1 g/L

Effect of Contact Time

To study the effect of contact time 0.5 g of 137.5 µm average particle size adsorbent is taken 30 ml of aqueous solution of initial methylene blue concentration 0.02 g/L, at known pH 6.8 and the shaking was provided for 35 minutes. The experiment was repeated for different time intervals like 1,2,3,4,5,6,7,8,9,10,15,20,25,30,35, minutes at constant agitation speed. After each interval of time the sample was filtered and analyzed to determine optimum contact time. The data obtained from the Adsorption of methylene blue on to WTP showed that a contact time of 25 minutes was sufficient to achieve equilibrium and the Adsorption did not change significantly with further increase in contact time.

Effect of Initial Concentration

To study the effect of methylene blue concentration 0.5 grams of Adsorbent is added to 30 ml of stock solution of methylene blue concentration 0.02 g/L and is kept shaking for optimum time then the procedure is repeated at pH 6.8 with 30 ml of stock solution with different initial concentrations 20, 40, 60, 80,100 mg/L keeping the agitation speed and room temperature constant then the sample was filtered and analyzed for methylene blue concentration.

Effect of Adsorbent Dosage

The effect of Adsorbent Dosage on the amount of methylene blue Adsorbed was obtained by agitating 30 ml of methylene blue solution of 0.02 g/L separately with 0.5, 1, 2, 3, 4 grams of Adsorbent at room temperature for optimum shaking time at constant agitation speed, maintaining the pH 6.8. The filtered solution of methylene blue was analyzed with the help of colorimeter to know the percentage Adsorption.

Effect of Average Particle Size

To study the effect of average particle size 30 ml of 20 g/L stock solution was added to 0.5 grams of known average particle size of adsorbent i.e., above 120 BSS; and it was kept until optimum time. The sample was filtered and analyzed for concentration of methylene blue. This experiment was repeated at constant agitation speed and room temperature with different particle sizes of Adsorbent from 137.5 µm to 302.5 µm mesh size.

Effect of pH

To determine the effect of pH the stock solutions of concentration 0.1 g/L with pH 6.8, acid was added in order to reduce pH value 2, 4, 6, and base was added to increase the pH up to 10. The acid used was freshly prepared hydrochloric acid and base was sodium hydroxide. After setting the pH of the ranges 2, 4, 6, 8, 10, 50 ml stock solution was pipette out in to each flask and 0.5 grams of WTP was added to it and allowed to undergo shaking for optimum time then the sample was analyzed for the percentage Absorption.

Adsorption Isotherms

The Adsorbent Isotherms indicate how the Adsorption molecules distribute between the liquid phase and the solid phase when the Adsorption process reaches an equilibrium state. The analysis of the isotherm data by fitting them to different isotherm models is an important step to find the suitable model that can be used for design purpose. Adsorption isotherm is basically to describe how solute interacts with adsorbents. The application of the isotherm equation is compared by judging the correlation coefficients R^2. Isotherms are calculated under existing conditions. These isotherms are useful for estimating the total amount of adsorbent needed to adsorb a required amount of Adsorbate from

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solution.Langmuir Isotherm: (C_{eq}/q_{eq}) = ($1/bq_{max}$) + ($1/ q_{max}$)C_{eq}

Freundlich Isotherm: log q_{eq} = log K_f + n log C_{eq}

Where, q_{eq} is the amount adsorbed per unit weight of adsorbents at equilibrium. C_{eq} is the equilibrium concentration of the Adsorbate (mg/L). q_{max} is equal to q_{eq} for a complete mono layer. n is the Freundlich constant. While b is the Langmuir constant and K_f is the Freundlich constant related to the Adsorption capacity.

Results and Discussions

Effect of Contact Time

The study had shown that for WTP the percentage of dye removal was high. The Adsorption data for the uptake of methylene blue verses contact time at initial concentration 0.02 g/L was shown. It indicated that the Adsorption of methylene blue increases with increase in contact time. The amount of dye uptake was found to occur in the first rapid phase (25 minutes) and their after the sorption rate was found to be constant. This is due to an increased number of vacant sites available at the initial stage and after a lapse of time, the remaining vacant surface sites are difficult to be occupied due to repulsive forces between the solute molecules on the solid and bulk phases.

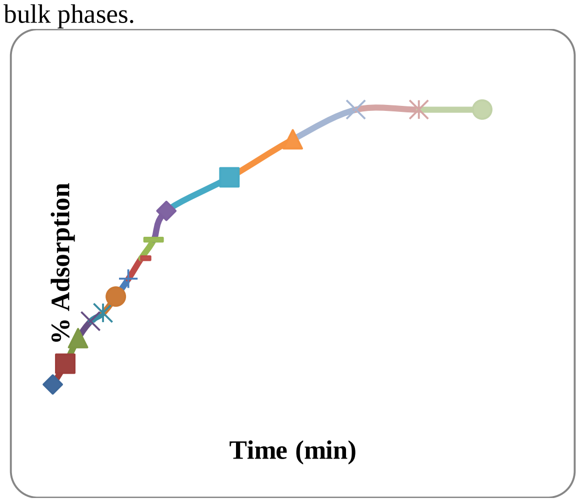

Figure-1 : Effect of Contact Time

Effect of Contact Time on % Adsorption of Methylene Blue on to WTP was studied over a shaking speed of 160 rpm and time of 35 minutes using 0.5 grams of WTP, 30 ml of 20 ppm of individual Methylene Blue solution concentration at pH 6.8, temperature 30ºC. The data obtained from the Adsorption of Methylene Blue on to WTP, showed that a constant time of 25 minutes was sufficient to achieve equilibrium and Adsorption did not change significantly with further increase of time.**3.2 Effect of Initial Concentration**

The study has shown that for WTP the percentage of dye removal was 70% high at initial concentration 20 ppm at a shaking speed of 160 rpm and time 25 minutes. The lowest dye removal 54.6% was observed for concentration of 100 ppm. This indicates that an increase in the dye concentration had caused the decrease in the percentage of dye removal, even though the amount of dye being adsorbed is increased. This is due to increase in the driving force of the concentration gradient as an increase in the initial dye concentration and decrease in percentage Adsorption may be attributed to a lack of sufficient surface area to accommodate much more adsorbate available in the solution.

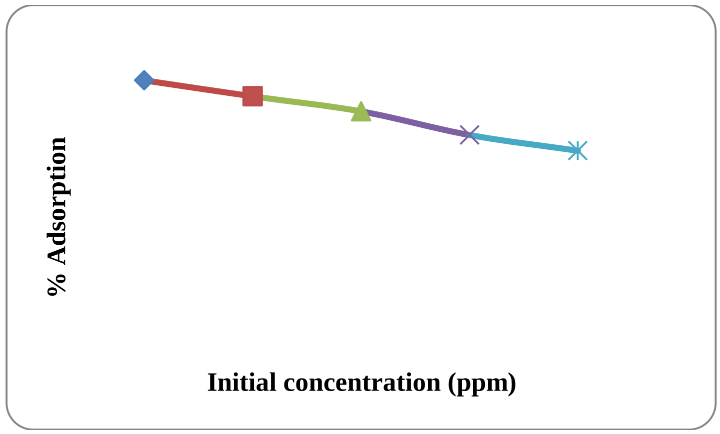

Figure-2: Effect of concentration of MB onto *WTP*

At lower concentrations almost all the adsorbate present in the solution could interact with the binding sites and thus % Adsorption higher than those higher at initial concentrations. At higher concentrations and lower Adsorption sites yield is due to the saturation of Adsorption of sites.

Effect of Adsorbent Dosage

From the figure 3, it was observed that, the amount of the dye adsorbed varied with varying adsorbent mass. The optical density decreased for an increase in Adsorbent mass from 0.5 to 4 grams, where as percentage color removal increased from 70 to 83 with an increase in adsorbent mass. The decrease in the optical density with increasing adsorbent mass is due to the split in the flux or the concentration gradient between solute concentration in the solution and the solute concentration in the surface of the adsorbent. The increase in the percentage of dye removal is due to increase in the surface area and availability of Adsorption site with increase in the Adsorbent dosage.

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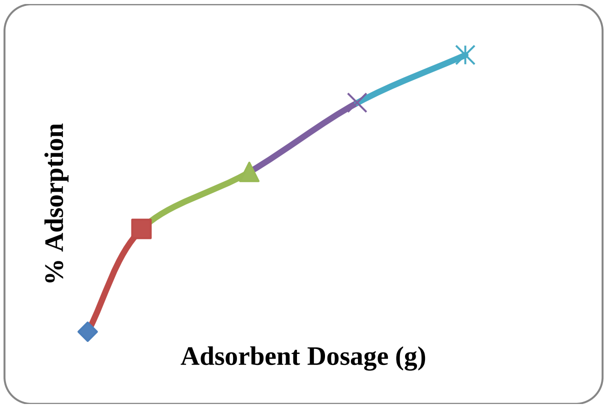

Figure-3: Effect of dosage on to Removal of Methylene Blue

Effect of Average Particle Size

The variation of the rate of Adsorption of the substance with different particle size of adsorbent is another factor influencing the rate of Adsorption. The present work was carried out at particle sizes 302.5, 230, 195, 165, 137.5µm, (BSS mesh sizes) and Adsorption of dye was monitored. Maximum Adsorption about 70% can be achieved at a particle size 137.5µm. This indicates that as the particle size increases the Adsorption rate decreases. High Adsorption with the smaller particle size is due to availability of more specific surface area on the Adsorbent.

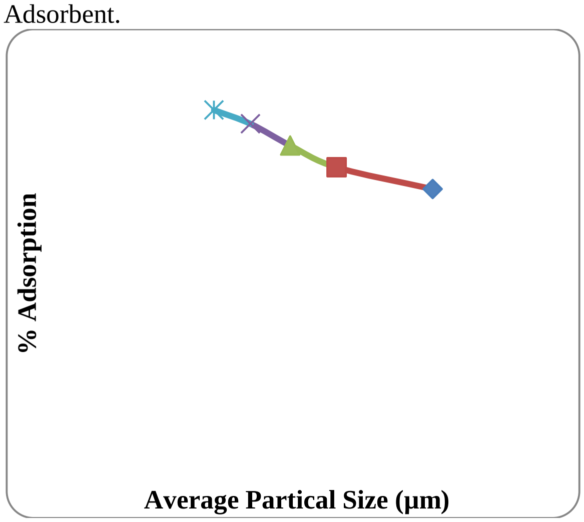

Figure-4: Effect of Average Particle size on Removal of Methylene Blue

Effect of pH

The study has shown that for the WTP the percentage of dye removal was 70% high at initial concentration 0.02 g/L at a shaking speed of 160 rpm and time 25 minutes. The lowest dye removal 54.60 % was observed for concentration of 0.1 g/L.

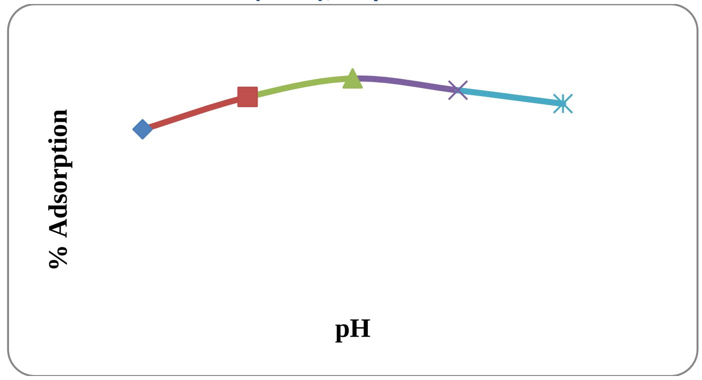

Figure-5: Effect of pH on Removal of Methylene Blue

The effect of pH on the percentage of the Methylene Blue is shown in figure 4.6 under various other fixed operating conditions. The initial pH of Adsorption medium is one of the most important parameters affecting the Adsorption process. It can be seen here that the percentage of dye removal was increased from 58.5 to 68.5 with an increase in the pH from 2 to 6, but there observed a decrease in dye removal from 68.5 to 62.1 the pH 6 to10. As expected, the adsorbent surface acidity and basicity are strong functions of pH of solutions. In this case the adsorbent surface acidity and adsorbent surface basicity might be responsible for such behavior in Adsorption and such surface properties can be expressed by the following equations;

Adsorbent-OH $\longrightarrow$ Adsorbent-O^-+H^+ (a)

Adsorbent-O^-+H^+ $\longrightarrow$ Adsorbent-OH_2^+ (b)

Equation (a) represents adsorbent surface basicity at higher pH. While equation (b) is represents adsorbent surface acidity at lower pH. Methylene Blue being an anionic dye, adsorbed onto the adsorbent surface effectually at lower pH values since the adsorbent surface attained at lower pH as per equation (b). As a result Adsorption of Methylene Blue on WTP was better at pH 6 to 7.

Langmuir Isotherm

Isotherms assume monolayer Adsorption onto a surface containing finite number of Adsorption sites of uniform strategies of Adsorption with no transmigration of Adsorbate in the plane of surface. The linear form of Langmuir Isotherm equation is given as;

$(C_{eq}/q_{eq}) = (1/bq_{max}) + (1/q_{max})C_{eq}$

Where q_{max} and b are Langmuir constants related to Adsorption capacity and rate of Adsorption respectively. A plot of C_{eq}/q_{eq} versus $1/C_{eq}$ for Methylene Blue to Adsorption onto WTP is presented in the figure. The Langmuir constants b = 0.028 and q_{max} = 5.785 are obtained from the graph. The R^2 value of 0.997 indicated that the Adsorption data of

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Methylene Blue onto WTP best fitted the Langmuir isotherm model

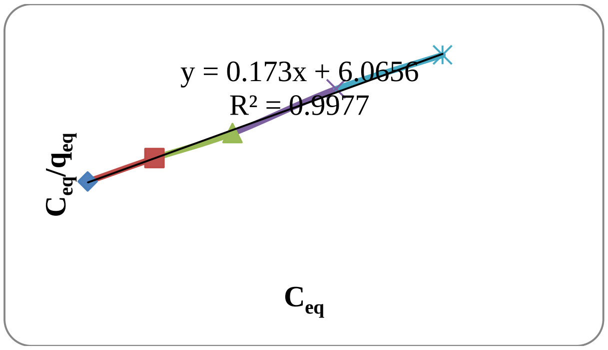

Figure-6: Langmuir Adsorption isotherm at 0.5 g /30 ml of Adsorbate concentration

Freundlich Isotherm

Unlike Langmuir Isotherm Freundlich isotherm assumes heterogeneous surface energies, in which the energy term in Langmuir equation varies as a function of the surface coverage, the linearized form of Freundlich isotherm can be written as:

$\log q_{eq} = \log K_f + n \log C_{eq}$

Where, K_f and n are Freundlich constants with n giving an indication of how favorable the Adsorption process. K_f can be defined as the Adsorption of distribution coefficient and represents the quality of dye adsorbed onto WTP for unit equilibrium concentration. The slope n ranging between 0 and 1 is a measure of Adsorption intensity or surface heterogeneity becoming more heterogeneous as its value coming closer to zero. From the figure 7, we obtain the slope n =0.6724 and intercept K_f =0.2652 & R^2 = 0.989.

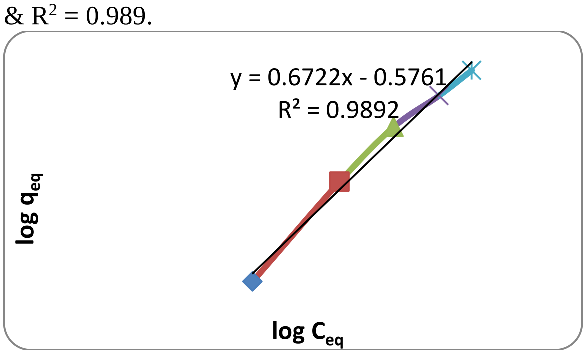

Figure-7: Freundlich Adsorption isotherm at 0.5 gm/30 ml of adsorbent

Table 1: Equilibrium constants for Adsorption of MB onto WTP

Isotherm	Constants	
Langmuir	b	0.028
	q_{max}	5.785
	R^2	0.9977
Freundlich	K_f	0.2652
	n	0.6724
	R^2	0.9892

The effect of isotherm can be used to predict whether a sorption system is favorable or unfavorable in batch process. Langmuir isotherm can be expressed in terms of a dimensionless constant separation factor of equilibrium parameter K_R which is defined by the $K_R = 1/(1+bCi)$

Table 2: Comparison of different adsorbent percent Adsorption with WTP adsorption capacity on Methylene Blue removal.

Types of adsorbent	**% Adsorption**
Activated carbon (coconut shell	75
Activated carbon (fruit peel)	73
Activated Carbon (palm kernel shell)	71
WTP (Present study)	**70**
Mesoporous material MCM-41	50
Pistachio shell	44
Zeolite material	40

From the above table it is clear that percentage Adsorption of Methylene Blue from aqueous solution having concentration 0.02 g/L on WTP is more than that compared to different adsorbents (Coconut shell, Archas sapota, Sugarcane Baggase, Tamarind Nut Shell) Adsorption capacity after undergoing a constant shaking of 25 minutes. So we carried out the whole Adsorption process with WTP as Adsorbent for highest concentration of Methylene Blue removal from aqueous solutions.

Conclusions

Analytically discussed and the following conclusions could be drawn from the study of removal of color from the solution of Methylene Blue using Adsorption technique.

1. The Adsorption performance is strongly affected by parameters such as initial concentration, pH, adsorbent dosage and adsorbent partical size.
2. The data obtained from the Adsorption of Methylene Blue on to WTP showed that a contact time of 25 minutes was sufficient to achieve equilibrium.
3. It was observed that 55.5 percentage Adsorption of Methylene Blue decreases with increase in the initial concentration of aqueous solution,
4. It was observed that percentage Adsorption of Methylene Blue decreases with increasing particle size of WTP .

5. The amount of adsorbate adsorbed increases with the increasing of adsorbent dose.
6. The experimental data gave good fit with Freundlich Isotherm & the Adsorption coefficient agreed well with condition of favorable adsorption.
7. The plot of pH versus percentage adsorption shows that more Adsorption take place at 6

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