Kinetic And Thermodynamic Study of the Removal of Textile Congo red Dye from Aqueous Solution by Adsorption onto Natural Adsorbent

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Abstract - Water pollution due to waste water disposal from industrial activities has been considered as one of the environmental issues in the world. Dye substances are major class of the pollutant. Dyes are toxic and concentration as low as 0.005 mg/L is visible which captures the attention of both public and authorities. Currently there are more than 100,000 commercial dyes with different chemical structures are used. The annual production of these dyes is assessed to be 7 x 10^5 – 1 x 10^6 tonnes per year . Dye molecules are synthetic complex aromatic with high stability and non biodegradable in nature condition . Cango red is one of the most common synthetic dyes used in the textile industry. Cango red (1-Naphthalenesulfonic acid, 3,3'-(4,4' biphenylenebis (azo) bis 4-amino) disodium salt) is a benezedene based dye known to metabolize to benzedene a known human carcinogen. It cause an allergic reaction is highly toxic and irritant properties to eye and skin contact Hence to study the removal of cango red dye from waste water by batch adsorption process. *Melia azedarach* leaf powder (Bakan neem , BNLP) use as a natural adsorbent for effective removal of cango red. The main parameters influence the adsorption of cango red as percentage recovery, initial metal concentration, effect of adsorbent dosage, effect of time and effect of pH was studied in batch process. The adsorption study werecarried out isothermally at different temperatures. Freudlich isotherm and Langmuir isotherm were used to describe the equilibrium data and the result were discussed in details.The kinetic data well described by the pseudo first order kinetic model. The thermodynamic parameters such as standard free energy change, entropy change and enthalpy change were studied for *melia azedarach* leaf powder (BNLP). This values showed that the adsorption of cango red on *melia azedarach* leaf powder (BNLP) was successfully employed for the removal of cango red from waste water and the techniques will be applicable at low cost and the percentage of removal of cango red on BNLP was very significant.

Keywords: Adsorption, *Melia azedarach*., Langmuir isotherm, Freundlich isotherm, Kinetic and Thermodynamics data.

I. INTRODUCTION

In developing country water pollution due to waste water disposal from industrial activities has been considered as one of the environmental issues in the world [1-6]. Organic & inorganicpollutants are discarging into water media by different industrial waste water gas stimulated worldwide attention because the harmful effect of pollutants affect to the environment and also human health. Dye substances are major class of the pollutant [7-8] .Various industries such as textile, paper, rubber,leather, cosmetic and food industries generated large volumes of dyecontaining effluents. Due to the chemical structures dyes are resistant to fading on exposure to light, water and many chemicals and therefore they are difficult to be decolourised once released into aquatic environment [9-10] .Dyes are toxic and concentration as low as 0.005 mg/L is visible which captures the attention of both public and authorities. Currently there are more than 100,000 commercial dyes with different chemical structures are used for printing and dying and a portion is discharged with waste water [11,12]. The annual production of these dyes is assessed to be 7 x 10^5 – 1 x 10^6 tonnes per year [13]. Dye molecules are synthetic complex aromatic with high stability and non biodegradable in nature condition [14]. Small quantity of dye contamination can be coloring the water body in large scale. This phenomenon can affect the light penetration to the water body and disrupt the photosynthesis process [15]. Because of the high environmental hazard, the removal of dye contaminant in the environment especially in a water

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body become seriously important. The various method including chemical, physical and biological treatment have been developed to remove the dye contaminant in waste water disposal [16]. Cango red is one of the most common synthetic dyes used in the textile industry. Cango red (1-Naphthalenesulfonic acid, 3,3'-(4,4' biphenylenebis (azo) bis 4-amino) disodium salt) is a benezedene based dye known to metabolize to benzedene a known human carcinogen. It cause an allergic reaction is highly toxic and irritant properties to eye and skin contact. At highlevel of contamination it induce same respiratory problems and even could be a carcinogenic agent to human.

Cango red removal from waste water of the textile industry is necessary to avoid environmental issues.Some of the most studied method for dye removal including coagulation, flocculation, membrane separation, oxidation or ozonation, bioremediation, electro-coagulation and adsorption [17]. The biological and physic-chemical process also used to remove the dye [18-20] Among these methods adsorption is the most potential, effective and reliable method for dye removal. At the major advantages of an adsorption treatment for the control of water pollution are less investment in terms of initial development cost, simple design, easy operations, free form generationof toxic substances and easy safe recovery of the adsorbent as well as adsorbate materials [22-25]. Recently the use of agricultural waste as natural adsorbent for dye was studied are relatively less expensive [26-28]. Agricultural waste, activated carbon prepared from agricultural waste and lignocelluloses materials are more effective and very inexpensive [29,30] such as coffee waste [31], date pits [32], coconut shell. [33].

Melia azedarach Linn. (Meliaceae, synonym Meliaorientalis Roam.), commonly known as "Persian Lilac", "Bakain" or "think" grows wild through out the sub-himalayan tract. It is cultivated in India and Pakistan for both ornamental and medicinal purposes [34-40]. Literature survey reveals that in many parts of the world preparations of *melia azedarach* are being used locally and systematically for curing many diseases [41-42]. The plant is considered as resolvent, deobstruent and alexipharmic. Locally flowers, leaves, fruits/berries and bark are used for curing many diseased skin conditions, such as eczema, ulcerative wounds, syphilitic ulcers, leprosy, scrofula etc. in the form of lotion, ointment or poultice. Systematically it is used as an emetic, cathartic, anthelmintic, antipyretic, expectorant and diuretic [43]. Commercially the oil of Melia azedarach is used in soap and cosmetic industries. Chemical composition reveals the presence of alkaloids, gum, resins, tannins, meliotannic acid, benzoic acid, vanillic acid, cinnamic acid, β-sitosterol, phenol, coumarin, teranor-triterpenoid, triterpenes, glycoside, bakayanin and margosine. [44].

Taxonomy

Subdivision : Angiospermae

Class : Dicotyledonae

Subclass : Polygonae

Series : Disciflorae

Order : Geraniales

Family : Meliaceae

Genus : Azedarach

Species : Melia

The objective of this work is to study the adsorption behavior of respect to cango red. The batch method was employed, parameters such as percentage recovery, , initial concentration of adsorbent, effect of addition of dose of adsorbent, effect of time, effect of temperature, Freundlich adsorption isotherm, Langmuir adsorption isotherm, Kinetic study and lso thermodynamic were studied.

II. MATERIALS AND METHODS

Materials

Adsorbent

The adsorbent selected for this study was ***Melia azedarach* leaf powder (Bakan neem , BNLP)** Which is locally available plant and was collected in Sangamner, Ahmednagar District of Maharashtra. The sample leaves were washed and dried in shadow, avoiding the direct sunlight on them. The dried leaves were grinded into powder and washed with distilled water and then filtered. The residue was treated with dilute solution of 0.1 N HCl and stirred continuously then wash with distilled water for the removal of acid. After that it was dried in oven at 95^0c for 6 hours. Then power it and use as a adsorbent.

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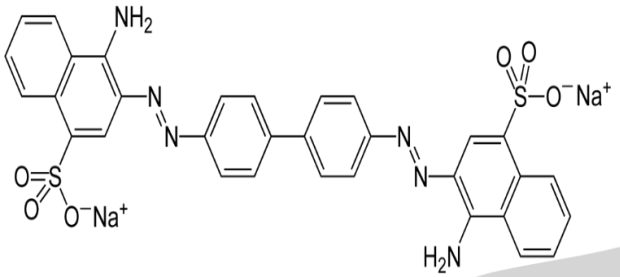
Adsorbate

Congo red, is an anionic azo dye having IUPAC name as 1-napthalenesulfonic acid, 3,3-(4,4-biphenylenebis(azo))bis(4-aminodisodium) . Its stock solution was prepared in double-distilled water. 1gm of Congo red was dissolved in 1L of double distilled water to obtain stock solution. Later it was diluted by using distilled water according to the concentration required and pH was adjusted by adding .1 M NaOH soln and .1 M HCl soln according to the conditions**.** All the test solutions were prepared by diluting the stock with double- distilled water.



Adsorption Study (Batch Process)

The dried powder of was ***melia azedarach*** **leaf powder (BNLP)** 1.5 gm was taken in stoppered bottle. In this bottle the stock solution of congo red with initial concentration of 10 mg/dm^3, 20 mg/dm^3, 30 mg/dm^3, 40 mg/dm^3, 50 mg/dm^{3}was added. The mixture were well stirred on a shaker at 100 rpm at the temperature 298 K, 303 K, 308 K and 313 K for 20, 40, 60, 80, 100 & 120 minutes until the equilibrium condition were reached. The content was filtered. The adsorbate and adsorbent were separated by filtration. The congo red filtrate in the aqueous solution after adsorption was measured by using pH values of the solution were determined by using pH 2 to 9 using pH meter. The percentage of adsorption was determined from initial and equilibrium concentration respectively.

III. RESULTS AND DISCUSSION

Effect of pH

Effect of pH of solution is very important in adsorption process of adsorbent . The pH of solution affect on surface of adsorbent. pH depends on the adsorbent and adsorbate nature. The effect of pH on the removal of congo red using *melia azedarach* leaf powder (BNLP) powder as an adsorbent. It was studied with the initial pH range from 2 to 9 it realates the initial pH of the solution and then the percentage of congo red. With increasing pH from 5 to 7 the percentage of congo red increases. Adsorption process is good at pH 6.5 . The uptake capacity of congo red ions is better for *melia azedarach* leaf powder (BNLP) powder

Effect of Adsorbent Dose

The effect of adsorbent on congo red removal was studied by batch adsorption process. The percentage of removal of congo red reaches about from 82.58 % . The dose required is near about 200 mg/ 25 ml for the initial concentration of 25 mg/L at pH 6.5.

Effect of contact time

The rate of removal of congo red, the effect on contact of congo red adsorption on *melia azedarach* leaf powder (BNLP) was studied. The percentage removal of congo red at different initial values. concentration of solution varies from 5,10,15,20,25,30,35,40,45,50 mg/L and batch experiments were carried out by taking 200 ml of this solution with dried 1.5 gm of adsorbent and the system is equilibrium by shaking the solution content at room temperature. The equilibrium of solution reaches in 7 hours. Final concentration of congo red was determined by spectrophotometrically. The removal of congo red was 82.58 %. The adsorption of congo red on *melia azedarach* leaf powder (BNLP) as a function of time was studied. As the concentration of *melia azedarach* leaf powder (BNLP) increases surface area of *melia azedarach* leaf powder (BNLP) was covered more & more and hence at the higher concentration of metalions capacity of congo red adsorbed in the surface of *melia azedarach* leaf powder (BNLP) is decreased due to unavailability of the surface area of *melia azedarach* leaf powder (BNLP). . It conclude that at lower concentration of congo red the percentage of adsorption is high because of the more active site of *melia azedarach* leaf powder (BNLP) is available for the adsorption. **Table 1**

Effect of Temperature

Temperature is very important factor for adsorption. Higher temperature increases the rate of the adsorbate and decrease in the viscosity of the solution. Change in the temperature changes the equilibrium capacity of the adsorbent for the particular adsorbate. A series of experiments were conducted at 298 K, 303 K, 315 K and 340 K to study the effect of temperature on the congo red time rate. for 20, 40, 60, 80, 100 & 120 minutes.

Freundlich Adsorption Isotherm

Freundlich plot for the adsorption of congo red with *melia azedarach* leaf powder (BNLP). shows that the values of adsorption intensity 1/n is less than 1, indicates the applicability of Freundlich adsorption. **Table 2**

Langmuir Adsorption Isotherm

The value of Q_0 of Langmuir adsorption isotherm found to be comparable with commercial activated carbon. Value of b lies between 0 to 1 it indicates that the adsorption is favorable. It indicate that the applicability of Langmuir adsorption isotherm . **Table 3**

Adsorption Kinetics

Adsorption rates of congo red on ions *melia azedarach* leaf powder (BNLP). was studied by first order kinetic rate equation. It is found that the initial concentration of congo red increases rate constant increases it indicate that the adsorption follow the first order kinetics. **Table 4.**

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Thermodynamic Parameters

Adsorption rate depends on temperature was investigated at298 K, 303 K, 315 K and 340 K. it conclude that increasing temperature mass of congo red per unit mass of the *melia azedarach* leaf powder (BNLP) was increased. At above temperature the change in Gibb's free energy, Change in enthalpy and Change in entropy was calculated. Change in Gibb's free energy shows negative value indicate that the adsorption of congo red on *melia azedarach* leaf powder (BNLP) is spontaneous and feasibility. The change in enthalpy value indicates endothermic nature of congo red on *melia azedarach* leaf powder (BNLP). The change in entropy shows positive value indicates increase randomness during the adsorption process of congo red on *melia azedarach* leaf powder (BNLP). **Table 5**

Table 1. Summary of % Recovery & Adsorbent Capacity Initial concentration 25 mg/L , *Melia azedarach* leaf powder (BNLP). dose 1 mg/L

Sr. No.	Adsorbent	Adsorbate	Final Conc (mg/L)	% Recovery	Q (mg/L)
1	*Melia azedarach* leaf powder (BNLP)	Congo Red	11.50 mg/L	82.58 %.	18.75

Table 2. Freundlich Adsorption Isotherm of Congo Red on BNLP

Sr. No.	Adsorbate	Concentration	Freundlich Constant	
			K	1/n
1	Congo red	10 mg/dm^3	4.9367	0.2563
		20 mg/ dm^3	5.0219	0.3979
		30 mg/ dm^3	5.9997	0.4888
		40 mg/ dm^3	6.1196	0.6021
		50 mg/ dm^3	8.4398	0.9613

Table 3. Langmuir Adsorption Isotherm of Congo Red on BNLP

Sr. No.	Adsorbate	Concentration	Langmuir Constant	
			K	1/n
1	Congo red	10 mg/dm^3	28.25	0.050
		20 mg/ dm^3	38.90	0.069
		30 mg/ dm^3	51.92	0.081
		40 mg/ dm^3	64.28	0.089
		50 mg/ dm^3	70.55	0.096

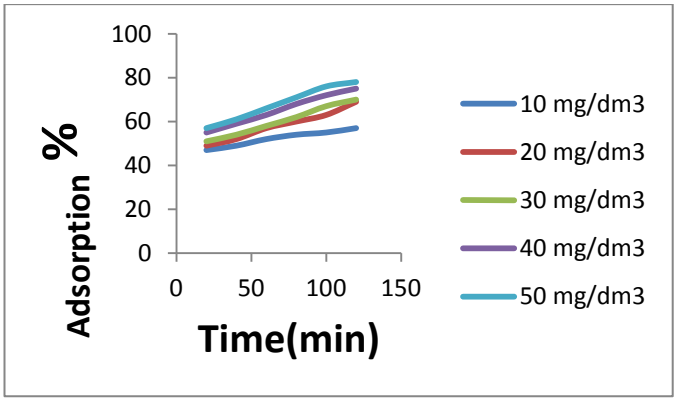
Table 4. Adsorption Kinetics of Congo Red on BNLP

Sr. No.	Adsorbate	Concentration	First order rate constant (K_1)
1	Congo red	10 mg/dm^3	2.99937 x 10^{-2}
		20 mg/ dm^3	5.99821 x 10^{-2}
		30 mg/ dm^3	7.01452 x 10^{-2}
		40 mg/ dm^3	8.11458 x 10^{-2}
		50 mg/ dm^3	9.92575 x 10^{-2}

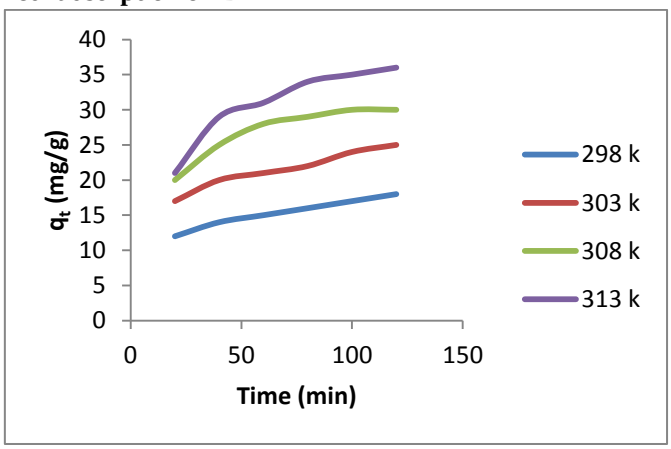
Table 5.Thermodynamic parameters for the adsorption of Congo Red on BNLP

Sr. No.	Adsorbate	T (^{0}K)	ΔG (KJ/mol)	ΔH (KJ/mol)	ΔS (KJ/mol)	R^2
1	Congo red	298	-19.84	46.27	0.5851	0.7993
		303	-37.25			
		315	-48.70			
		340	-52.93			

Graph 1 : Effect of contact time and initial concentration of congo red adsorption on MNLP Dose 1.5vg/dm^3(pH=6.5 at 313 K)



Graph 2 : Effect of temperature on adsorption of congo red adsorption on BNLP



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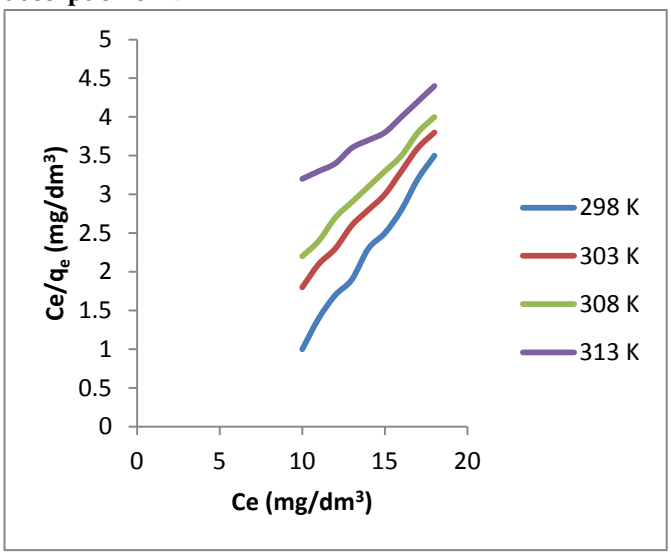
Graph 3 : Langmuir Adsorption isotherm of congo red adsorption on BNLP



IV. CONCLUSION

From above study it will be conclude that

***** BNLP was used as a adsorbent for removal of congo red dye. It is good adsorbent. Adsorption process is rapid at the starting and a becomes slow at the standard stage. It dependent on initial concentration of congo red and also time for adsorption.

* BNLP does increased percentage of adsorption also increased.

* This adsorption is good agreement with Freundlich adsorption isotherm and also for Langmuir adsorption isotherm .

* Adsorption process is good at pH 6.5. The uptake capacity of crystal violet is better for BNLP

* Temperature effect shows that with increasing temperature capacity of adsorption increases.

* BNLP could be exploited for commercial applicable.

* The cost of adsorbent is very low & is easily available.

* The adsorbent BNLP can be deposed safely.

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