

Removal of DNB-106 dye from dye wastewater using *Athi* tree leaf powder carbon (*Ficus racemosa*) by adsorption

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ABSTRACT

Athi tree leaves (*Ficus religiosa*) powder carbon was used as an adsorbent for the removal of DNB-106 dye from an aqueous solution. The studies were carried out under various experimental conditions such as agitation time, dye concentration, adsorption dose, and pH to assess the potentiality of *Athi* leaves powder carbon (ATC) for the removal of DNB-106 dye from wastewater. A greater percentage of dye removal was observed with decrease in the initial concentration of dye and increase in amount of adsorbent. The adsorption of dye on ATC was found to follow a gradual process. Equilibrium isotherms were analysed by the Langmuir models of adsorption and were applicable with maximum monolayer adsorption capacity of 4.5mgg⁻¹. The dimensionless factor, R_L of the DNB-106 dye, ATC isotherm revealed that the adsorption process is favourable in nature.

Key words: Adsorption, DNB-106, ATC, isotherm, *Ficus racemosa*.

INTRODUCTION

The greatest environmental polluter is the chemical industry, of which only a relatively small part pertains to the organic colourants industry (3-4%)¹. However, the dye industry is a multi product industry, and the costs of existing dyes and the planned costs of dyes in development (using and administration) must be determined on a relatively small sales basis. This result in further products that can satisfy the necessary financial criteria, with a consequent decreased introduction of products with improved technical properties and/or more acceptable environmental advantages².

Untreated textile effluents are highly toxic as they contain a large number of metal complex dyes. The effluents discharged from various textile and dyeing industries are hazardous coloured effluents. The greater environmental awareness in

both the public and regulator sphere in recent years has necessitated for effective treatment of industrial effluent³. As such, there has been a great deal of research work into finding cost-effective methods and for the removal of contaminants from wastewater. Interest in this area is exemplified by publication of several reviews worldwide⁴⁻¹⁰.

In recent past considerable attention has been devoted to the study of removal of dyes from wastewater by adsorption using agricultural products and by-products¹¹⁻¹³. Natural materials that are available in large quantities from agricultural operations may have potential to be used as low cost adsorbents, widely available and are environmental friendly¹⁴⁻¹⁵. The present study deals with the adsorption efficiency of *Athi* tree (*Ficus racemosa*) of family Moraceae, *Athi* leaves powder carbon (ATC) for the removal of DNB-106 dye from aqueous systems. The properties of adsorbates and

adsorbents are quite specific and depend upon their constituents. The constituents of adsorbents are mainly responsible for the removal of any particular pollutants from wastewater¹⁶.

Preparation and characterization of the adsorbent and dye

All chemicals were of analytical reagent grade and supplied by BDH India. The materials were used after proper purification using recommended methods¹⁷. Adsorption behaviour depends on the method of preparation and the prior treatment of the adsorbent. The adsorbent sample once prepared was stored and was used throughout the series. Athi Tree (Athi Maram - in Tamil) Leaves (*Ficus racemosa*) of family Moraceae were collected from a local village temple and then dried at a

temperature of 60–80°C. This was powdered by grinding in a mechanical blender and added in small portion to 2.0 l of 98% sulphuric acid during 10 h and the resulting mixture was kept for 2 days at room temperature followed by refluxing for 6 h in fume hood. Cool in ice bath and the reaction mixture was poured onto cold water and filtered. The obtained dehydrated leaves was heated in an open oven at 120°C for 24h followed by immersed in 5% sodium bicarbonate overnight to remove any remaining acid. The obtained carbon was then washed with distilled water until pH of the activated carbon reached 6, dried in an oven at 150°C for 48 h in the absence of oxygen and sieved to the particles of 200-800 micron (μ) sizes and stored in plastic bottle for further use.

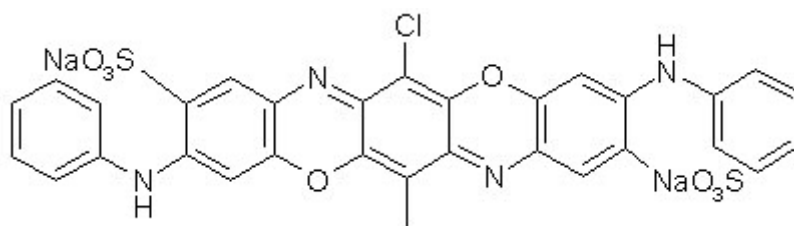


Fig. Chemical Structure of DNB - 106

RESULTS AND DISCUSSION

Adsorbates

The dye in the present study was selected from the list of dyes normally used in India, e.g. methylene blue, malachite green, crystal violet, rhodamine B, and Grey BL in paper industry, synthetic fibre and textiles industries. Synthetic wastewater was prepared by dissolving DNB-106 dye. The chemical nature of the adsorbate has profound effect on the adsorption characteristics.

Effect of contact time and concentration

The adsorbate concentration and contact time between adsorbent and adsorbate species play a significant role in the process of removal of pollutants from water and wastewater by adsorption at a particular temperature and pH. A rapid uptake of pollutants (dye) and establishment of equilibrium in a short period signifies the efficiency of the adsorbent for its use in wastewater treatment. In addition to this, the contact time is one of the factors

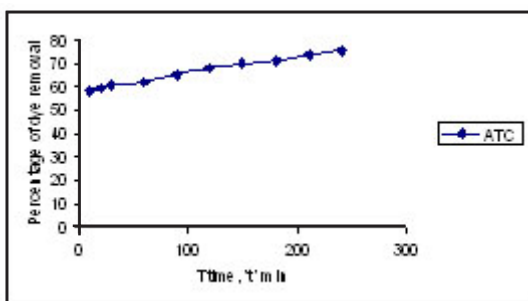
for the development of surface charges at the solid solution interface. The initial concentration of adsorbate also plays an important role as a given mass of the adsorbent can adsorb only a fixed amount of the solute. The more concentrated the solution or effluent, the smaller is the volume of effluent that a given mass of adsorbent can purify. In high concentration range, the fractional adsorption is low. Batch adsorption experiments were carried out by shaking the adsorbent with an aqueous solution of the dye of desired concentration in corning glass bottles at $27 \pm 1^\circ\text{C}$ in a water thermostat. The initial pH of the dye solutions was maintained at 7.2. The finding of the present investigation is given in Table 1. The removal of the dye DNB-106 by ATC increased from 2.06 to 3.61 mgg^{-1} by increasing the concentration of the dye from 6 to 12 mg L^{-1} at temperature $27 \pm 1^\circ\text{C}$ and pH 7.2. However, the percentage of the dye adsorbed by ATC decreases from 69.02 to 62.01% for DNB-106 by increasing the concentration of the dye from 40 to 100 mgL^{-1} .

Table 1: Amount, percentage and equilibrium time of DNB-106 dye adsorped (pH 7.0; Temperature 27°C)

Conc. of DNB-106 (Mg/l)	Amount adsorbed (mg/g)	% removal	Equilibrium time (min)
40	2.06	69.02	15
60	2.51	67.40	15
80	3.10	65.01	15
100	3.61	62.01	15

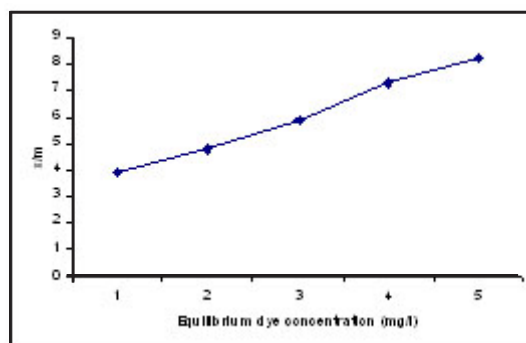
It is clear from Fig. 1 that the extent of adsorption is rapid in the initial stages and becomes slow in later stages till saturation is attained. This is obvious from the fact that a large number of surface sites are available for adsorption at the initial stages and after a lapse of time, the remaining surface sites are difficult to be occupied because of repulsion between the solute molecules of the solid and bulk phases. The time required to attain equilibrium in the case of adsorption of dyes is 15 min. It can be seen from the figure that the equilibrium period remains unaltered with the change of initial concentration of the dye solution.

A series of contact experiments were undertaken for varying initial dye concentrations namely 40, 60, 80 and 100mgL⁻¹. For low concentrations, there was a rapid uptake of dye due to surface mass transfer¹⁸. The time variation curves for adsorption are simple, smooth and continuous, indicating the formation of monolayer coverage on the surface of adsorbent.

**Fig. 1 Effect of concentration of DNB-106 dye by ATC (Temperature 300K; pH 7.0)**

Adsorption isotherm

The purpose of the adsorption isotherms is to relate the adsorbate concentration in the bulk and the adsorbed amount at the interface. To study the adsorption isotherms, 100 mg of adsorbent (ATC) was taken with 50mL of an aqueous solution of the dye at different concentrations (10–100mg L⁻¹) in a water bath until the equilibrium is established in each bottle. The experiments were carried out at 27°C.

**Fig. 2 Equilibrium adsorption of DNB-106 dye**

The Langmuir isotherm is valid for monolayer adsorption onto a surface containing a finite number of identical sites. The model assumes uniform energies of adsorption onto the surface and no transmigration of adsorbate in the plane of surface. The experimental data for the uptake of the dye DNB-106 dye by adsorbent (Athi leaves powder carbon) have been correlated with the rearranged Langmuir's model of adsorption in the present investigation:

$$\frac{C_e}{q_e} = \frac{C_e}{Q^0} + \frac{1}{Q^0 b}$$

where C_e is the equilibrium concentration (mg L⁻¹) of the dye, q_e is the amount (mg g⁻¹) of the dye adsorbed at equilibrium and Q^0 Fig. 3. Langmuir plot for the adsorption of DNB-106 on ATC at a temperature of 27°C and b are the Langmuir constants related to adsorption capacity and energy of the adsorption, respectively. The plot of C_e/q_e versus C_e at 27°C for the DNB-106 (Fig. 3) is found to be linear, suggesting the applicability of Langmuir isotherm.

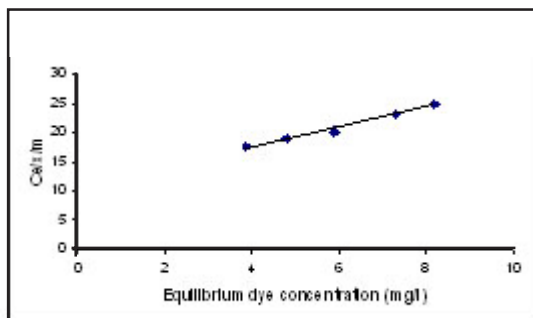


Fig. 3 Langmuir plot of Grey BL dye

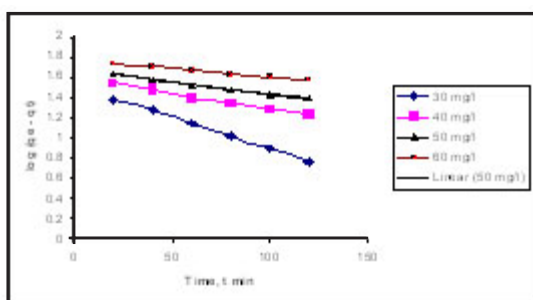


Fig. 4. Lagergren plot for the adsorption of DNB-106 dye on ATC at (Concentration 10mg L-1; pH: 7.2).

The Fig. 5 shows the plot of the log values of amount of dye adsorbed per weight of carbon against the log values of equilibrium dye concentration (C_e). The straight-line nature of the plot indicates that the process followed were Freundlich type of adsorption. The 'k' values of Freundlich adsorption equations were obtained for carbon from the intercept of straight line and $\log 1/n$ value, i.e., sorption intensities are calculated from the slope.

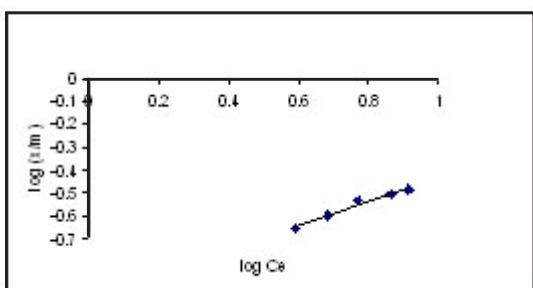


Fig. 5 Freundlich plot of DNB-106 dye

The essential characteristics of a Langmuir isotherm can be described by a separation factor or equilibrium parameters R_L which is defined by

$$R_L = \frac{1}{1 + bC_0}$$

where C_0 is the critical concentration (mg L^{-1}) and b is the Langmuir constant. The value of R_L for the adsorption of DNB-106 on ATC leaves powder indicates that the adsorption process is very favourable because R_L value lies between 0 and 1.

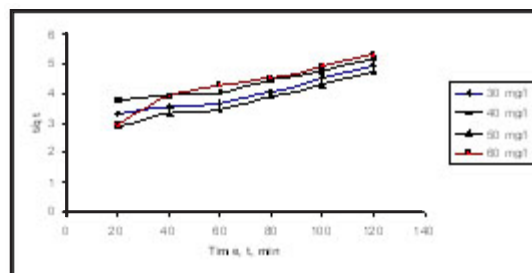


Fig. 6 Pseudo-second order sorption kinetics of DNB-106 dye by ATC at different DNB-106 concentrations (adsorbent dose 0.1 g, Temperature 27°C).

Intra-particle diffusion

The plots of qt vs $t^{0.5}$ (Fig. 7) were found to be linear with regression coefficients of 0.90-0.99. The intraparticle diffusion rate constant k_i was in the range of $(1.21 \text{ to } 14.17 \times 10^{-2} \text{ mg g}^{-1} \text{ min}^{-0.5})$. The linearity of the plots demonstrated that intra particle diffusion played a significant role in the uptake of

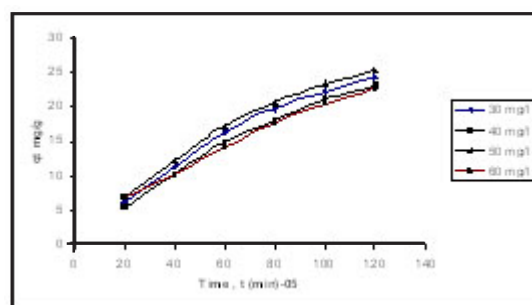


Fig. 7 Plots of qt (mg g^{-1}) vs $t^{0.5}$ for evaluating intra particle diffusion rate constant.

the dye by Athi leaf powder carbon. This also confirms that adsorption of the dye on the adsorbent was a multi step process, involving adsorption on the external surface and diffusion into the interior.

Effect of pH

The removal of pollutants from water and wastewater by adsorption is greatly influenced by the pH of solution, which affects the nature of the surface charge of the adsorbent as well as the extent of ionization and speciation of the aqueous adsorbate species and consequently the rate of adsorption. A change of pH affects the adsorptive process through dissociation of functional groups on the adsorbate and adsorbent¹⁸.

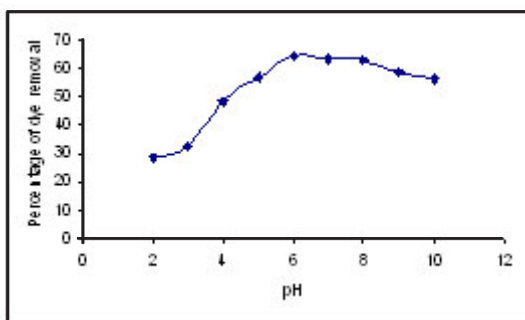


Fig.8. Effect of pH on the removal of DNB-106 dye by Athi leaves powder (concentration: 30mg L⁻¹; temperature: 27±1°C).

The adsorption of DNB-106 dye increases with the increase of pH of the solution. The variation in the amount of adsorbed dye with respect to pH of

the medium has been shown graphically (Fig. 8). The maximum removal was noted at pH 7.2. With an increase of pH from 4.2 to 7.2, the uptake of DNB-106 increases from 28.45 % to 63.99.11% which is close to the satisfactory level by Arasu leaves powder at a temperature 27±1°C and concentration 30mgL⁻¹ of the dye. It appears from the results obtained in the present investigation that in the alkaline medium the negatively charged species starts dominating and the surface tends to acquire a negative charge. As the adsorbent surface is negatively charged, the increasing electrostatic attraction between positively charged adsorbate species and negatively charged adsorbent particles would lead to increased adsorption of the dye.

CONCLUSION

Athi tree (*Ficus racemosa*) leaves powder carbon shows significant adsorption capacity for DNB-106 dye under suitable experimental conditions and hence will serve as an useful adsorbent. The Athi leaves is abundantly available and inexpensive too. Its binding capacity of basic dye is appreciably high.

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REFERENCES

1. Clarke, E.A. and Steinle, D., *Rev. Prog. Coloration*, 1995, **25**, 1.
2. Slokar, Y.M., and Majcen Le Marechal, A. *Dyes and pigments*, 1998, **37**(4), 385-396.
3. Abdel-Ghani, N.T., Hefny, M., El-Chaghaby, G.A.F. *Int. J. Environ. Sci. Technol.* 2007, **4**(1), 67-73.
4. Forgacs, E., Cserhati, T. and Oros, G. *Environ. Int.* 2004, **30**, 953-971.
5. Robinson, T., McMullan, G., Marchant, R., Nigam, P. *Bioresour. Technol.* 2001, **77**, 247-255.
6. Crini, G. *Bioresour. Technol.* 2006, **97**, 1061-1085.
7. Vandeviere, P.C., Bianchi, R., Verstrete, W. *J. Chem. Technol. Biotechnol.* 1998, **72**, 289-302.
8. Hernandez-Ramirez, O., Holmes, S.M. *J.*

- Mater. Chem.* 2008, **18**, 2751–2761.
9. Gupta, V.K. and Suhas. *J. Envir. Manag.*, 2009: (in press)
 10. Ayhan Demirbas, *J. Haz. Mat.*, 2009 (in press)
 11. Meyer, V., Carlsson, F.H.H., Oellermann, R.A. *Water Sci. Technol.* 1992, **26**, 1205–1211.
 12. Allen, S.J., Mckay, G., Khader, H.K.Y. *J. Colloid Interf. Sci.* 1998, **126**, 517–524.
 13. Hameed, B.H., Ahmad, A.L., Latiff, K.N.A. *Dyes Pigments* 2007, **75**, 143–149.
 14. Gong, R., Sun, Y., Chen, J., Liu, H., Yang, C. *Dyes Pigments* 2005, **27**, 175–181.
 15. Viorica Dulman, Simona Maria, Cucu-Man. *J. Haz. Mat.*, 2009, **162**, 1457-1464.
 16. Khattri, S.D., Singh, M.K. *Indian J. Chem. Technol.* 1999, **6**(2), 112–116.
 17. Vogel, A. A Text Book of Quantitative Inorganic Analysis, 15th ed., Longman, London, 2006.
 18. Annadurai, G., Juang, R.S. Lee, D. *J. Hazard. Mater.* 2002, **B92**, 263–274.